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ORNL-2989
UC-41 - Health and Safety

PLUTONIUM RELEASE INCIDENT OF NOVEMBER 20, 1959

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OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

Printed in USA. Price $2.00. Available from the
Office of Technical Services
Department of Commerce
Washington 25, D.C.

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Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

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DATE ISSUED
FEB 1 1961
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ABSTRACT

A nonnuclear explosion involving an evaporator occurred in a shielded cell in the Radiochemical Processing Pilot Plant at Oak Ridge National Laboratory on Nov. 20, 1959. Plutonium was released from the processing cell, probably as an aerosol of fine particles of plutonium oxide, via three principal routes:

(1) Cell ventilation system: ~1.5 g completely removed from the air stream by roughing and absolute filters.

(2) Cell 6 door, blown open (but not off) to the outside: ~600 mg spread rapidly over limited area south and east of building; Graphite Reactor Building most contaminated.

(3) Pipe passages and service openings from cell 6 to the interior of Bldg. 3019: ~70 mg; required decontamination over several months.

It is probable that this evaporator system had accumulated ~1100 g of nitric acid-insoluble plutonium in the steam stripper packing; the explosion released an estimated 150 g inside cell 6, with about 135 g in the evaporator subcell and about 15 g in the larger main cell.

No radioactive material was released from the ventilation stacks; no contamination of grounds and facilities occurred outside of a relatively small area of Oak Ridge National Laboratory immediately adjacent to the explosion.

No one was injured by the explosion, and no one received more than 2% of a lifetime body burden of plutonium or an overexposure to sources of ionizing radiation either at the time of the incident or during subsequent cleanup operations. The explosion is considered to be the result of rapid reaction of nitrated organic compounds formed by inadvertent nitration of about 14 liters of a proprietary decontaminating reagent.

In cleanup the contamination was bonded to the nearby street and building surfaces with tar, paint, roofing compound, or masonry sealer, as appropriate to the surface. Decontamination of the interior of the pilot plant building, except the processing cells, was 95% complete on Sept. 1, 1960. Decontamination of the processing cells was delayed 8 months until building modifications could be made to improve containment. Modifications to the pilot plant have been proposed which will preclude discharge into the Laboratory area and its environment of concentrations or amounts of radioactive materials that would be injurious to health or interfere with other Laboratory programs.

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1.0 SUMMARY

On November 20, 1959, at about 11:00 p. m., a nonnuclear explosion involving an evaporator occurred in a shielded cell in the Radiochemical Processing Pilot Plant at Oak Ridge National Laboratory. Plutonium released from the processing cell contaminated areas in the pilot plant building and nearby streets and building surfaces. No radioactivity was released to areas outside of the Laboratory, no one was injured by the explosion, and no one received a significant fraction of a lifetime body burden of plutonium or an overexposure to sources of ionizing radiation either at the time of the incident or during subsequent cleanup operations.

The explosion is considered the result of rapid reaction of nitrated organic compounds formed by the nitration of 14 liters of Turco Decon 4501, a proprietary decontaminating reagent.

The cell in which the explosion occurred is one of four cells containing equipment for the solvent extraction processing of highly irradiated nuclear fuels. The evaporator, contained in a subcell that is walled off by concrete blocks within the cell, consisted of a steam stripper, vapor separator, a natural-convection evaporator loop, and connecting piping.

About one month preceding the incident, after several processing campaigns with highly irradiated uranium had been completed, a decontamination program was begun. One objective of this program was to decontaminate the evaporator subcell which was emanating greater than 100 r/hr of penetrating radiation.

After two series of decontamination treatments did not decrease the radiation level in the evaporator subcell, Turco Decon 4501 was added to the evaporator and heated 2 hr. The evaporator was drained through a remotely operated product outlet valve because the valve normally intended for draining was located in the 100-r/hr radiation zone; this left 13.9 liters of organic compounds in the evaporator. Next, 270 liters of 4 M nitric acid was added to the evaporator, omitting the normal water flush. The heat exchanger steam was turned on, providing sufficient heat to concentrate the nitric acid and allow complete reaction between the acid and organic compounds. As the acid became concentrated, the temperature increased steadily until the nitro compounds, just formed, ignited and exploded.

The explosion was a minor chemical explosion and physical damage was confined to the equipment in the evaporator subcell, a few pipes that were connected to this equipment, and the unmortared concrete block shielding walls in front of the evaporator subcell and the processing cell door. The cell ventilation filters retained the radioactivity with a high degree of efficiency, but the momentary increase in cell air pressure caused airborne activity to be transported from the processing cell via pipe access holes to other areas in the building and via a cell door to the outside of the building.

In cleanup the plutonium was fixed to contaminated outdoor surfaces with either paint, tar, roofing compound, or masonry sealer, as appropriate to the surface.

Decontamination of all contaminated areas in Bldg. 3019 involved vacuuming and sponging all surfaces to remove easily transferable contamination and scrubbing with detergent solutions to remove the remainder of the transferable activity. The most difficult part of the decontamination was the removal of nontransferable contamination, >800,000 d/m/100 sq cm in some places, that had become fixed to concrete, metal, and painted surfaces. It was generally necessary to mechanically remove a layer of the contaminated surfaces to remove this activity.

It was decided that an area in Bldg. 3019 would be classified as a "noncontamination zone" only when a survey involving one smear and four measurements with a gas-purged proportional α counter per square meter of projected surface area resulted in (1) transferable activity (smear results) of no more than 3 d/m/100 sq cm, average, with no single smear indicating more than 30 d/m/100 sq cm and (2) nontransferable activity of 30 d/m/100 sq cm, average, with no single measurement of more than 300 d/m/100 sq cm. All building areas except the processing cells were either decontaminated to these specifications or, if this was not feasible, were decontaminated to within ten times these specifications and the surfaces were color coded with red or orange paint and then covered with either paint or concrete. Areas with painted-in activity are designated as "controlled zones" and in such areas, when the bright color shows, the surfaces will be repainted.

Since this incident, all operations at the Laboratory involving significant quantities of radioactive materials have been examined to ensure

that the maximum credible accident will be contained and that two lines of defense are present to prevent escape of radioactive materials via waste streams. The application of these containment criteria to Bldg. 3019 indicates the need for structural changes to the building, modifications to the off-gas and ventilation systems (including improved instrumentation), and additional equipment for fire and explosion protection.

The release of plutonium from the process equipment and from the containing shielded cell was the only significant result of the explosion, which fortunately occurred at a time when the ORNL population was low and a full week end was available for decontamination and immobilization of the radioactive material.

The explosion resulted from a combination of errors. First, an error in judgment was made in permitting a proprietary agent containing easily nitrated phenol in the process equipment. Since the incident the manufacturer has removed phenol from this highly effective formulation with no apparent loss in decontamination ability. Second, an inadvertent error in shift operating instructions caused nitric acid to be added directly to a small quantity of the decontaminating agent that was left in the equipment because of omission of the normal water wash. Last, the strong nitric acid--organic mixture was boiled for several hours, a step not normally included, which resulted in formation of the explosive, nitrophenol or picric acid.

2.0 INTRODUCTION

On Nov. 20, 1959, at Oak Ridge National Laboratory, the Radiochemical Processing Pilot Plant (Bldg. 3019), the X-10 Graphite Reactor (Bldg. 3001), and nearby streets and building surfaces became contaminated by plutonium that was released from the processing cells of the pilot plant because of an explosion in an intercycle evaporator that was being decontaminated. A preliminary report of this incident* was made to the Atomic Energy Commission at a time when the cause of the explosion, the extent of damage to equipment, and the extent of the contamination were not completely known. This report describes the explosion, its cause, the extent of the contamination, the decontamination of the Radiochemical Processing Pilot Plant and nearby buildings and ground, and the radioactivity containment program that has been proposed to preclude any future release of radioactivity from the pilot plant.

*Summarized in Nuclear Safety, Vol. 1, No. 3, pp. 78-80 (March, 1960).

Acknowledgment is made to J. R. Parrott, Problem Leader for Bldg. 3019 Power Reactor Fuel Processing operations, for information on the explosion and Bldg. 3019 decontamination; to W. N. Tillery, of the Oak Ridge National Laboratory Photographic Section, for the photographic record of the post-explosion condition of the evaporator and processing cells; and to the Oak Ridge National Laboratory Inspection Engineering Division for the mechanical and metallurgical analyses of the explosion.

3.0 THE EVAPORATOR EXPLOSION

On Nov. 20, 1959, at about 11:00 p. m., a nonnuclear explosion involving an evaporator occurred in a shielded cell in the Radiochemical Processing Pilot Plant at Oak Ridge National Laboratory, Oak Ridge, Tenn. Plutonium released from the processing cell contaminated areas in the pilot plant building and nearby streets and building surfaces. No radioactivity was released from Oak Ridge National Laboratory, no one was injured by the explosion, and no one received a significant fraction of a lifetime body burden of plutonium or an overexposure to sources of ionizing radiation either at the time of the incident or during subsequent cleanup operations.

3.1 Location of the Explosion

Oak Ridge National Laboratory is located 7 miles southwest of the city of Oak Ridge, Tenn., and 20 miles west of Knoxville, Tenn.

The intercycle evaporator that was ruptured is in a shielded subcell in cell 6 of the Radiochemical Processing Pilot Plant, Bldg. 3019 (Fig. 3.1). Figures 3.2, 3.3, 3.4 are isometric, plan, and sectional views of the building. The Analytical Chemistry Division occupies areas at the west end of the main floor—analytical hot cells and laboratories, a counting room, and offices. The Chemical Technology Division occupies areas at the east end of the main floor—cells and supporting areas required for radiochemical processing.

Highly radioactive processing equipment is located in one 11 by 20 by 27 ft deep and six 20 by 20 by 27 ft deep concrete cells, two of which, cells 6 and 7, are combined to form one large cell area. The six cell areas, cells 1 through 5 and the cells 6 and 7 combination, have 5-ft-thick walls on three sides and a 4-ft-thick wall on the outer side of the building. Entrances to cells 1 through 5 are 8 ft below grade level at the bottom of stairways which

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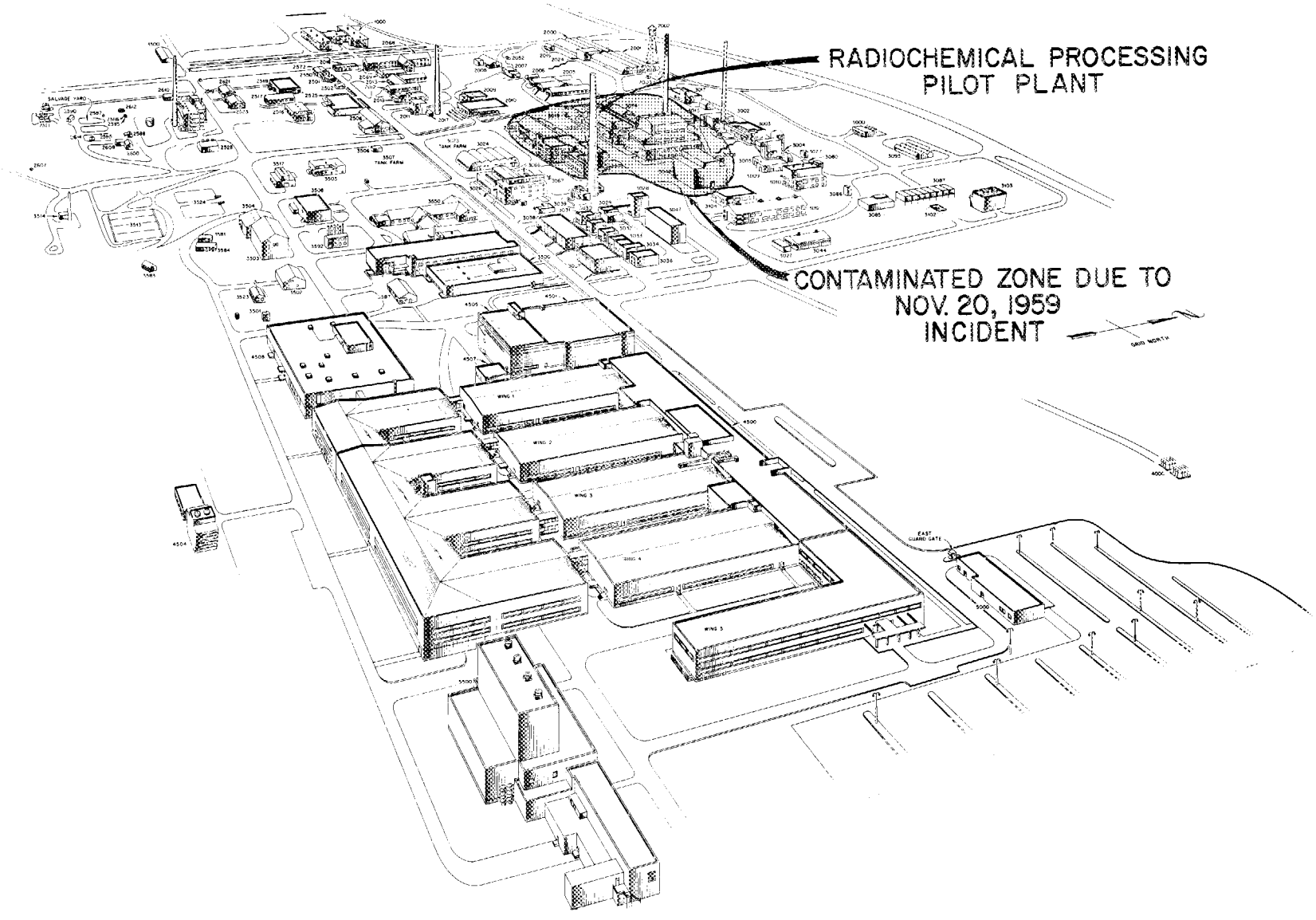

Fig. 3.1. Oak Ridge National Laboratory, X-10 Site.

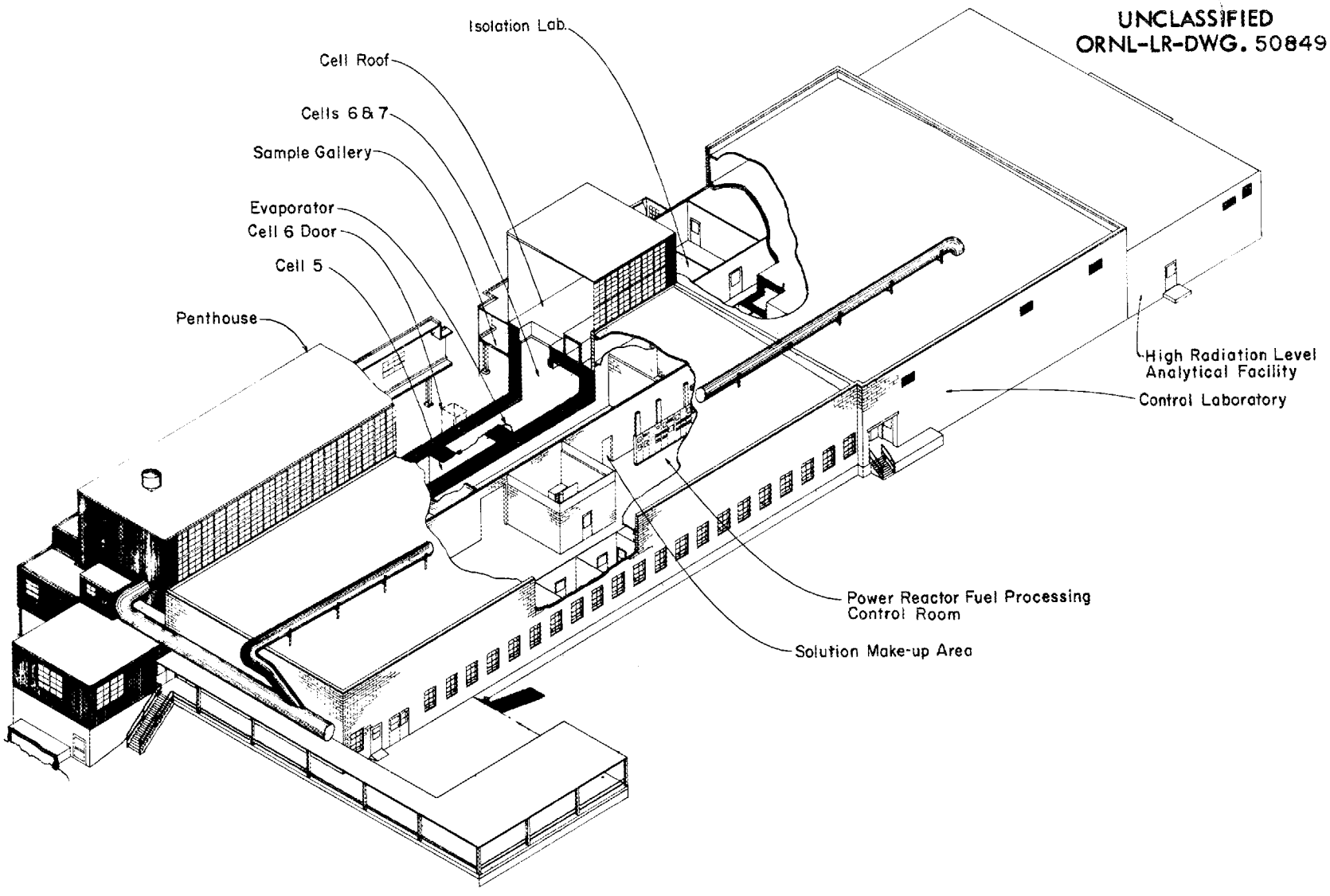

Fig. 3.2. Radiochemical Processing Pilot Plant.

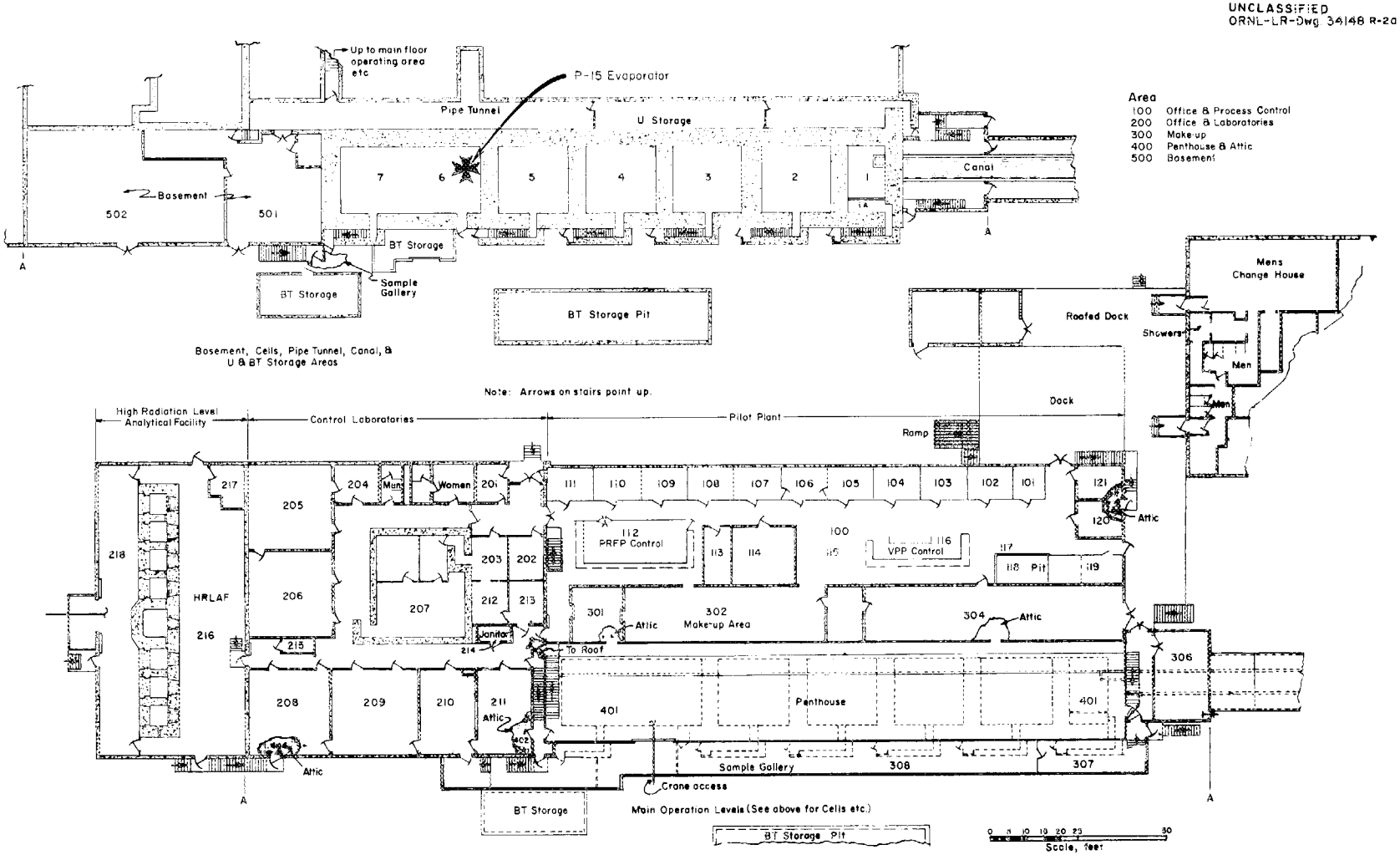

Fig. 3.3. Plan of Radiochemical Processing Pilot Plant.

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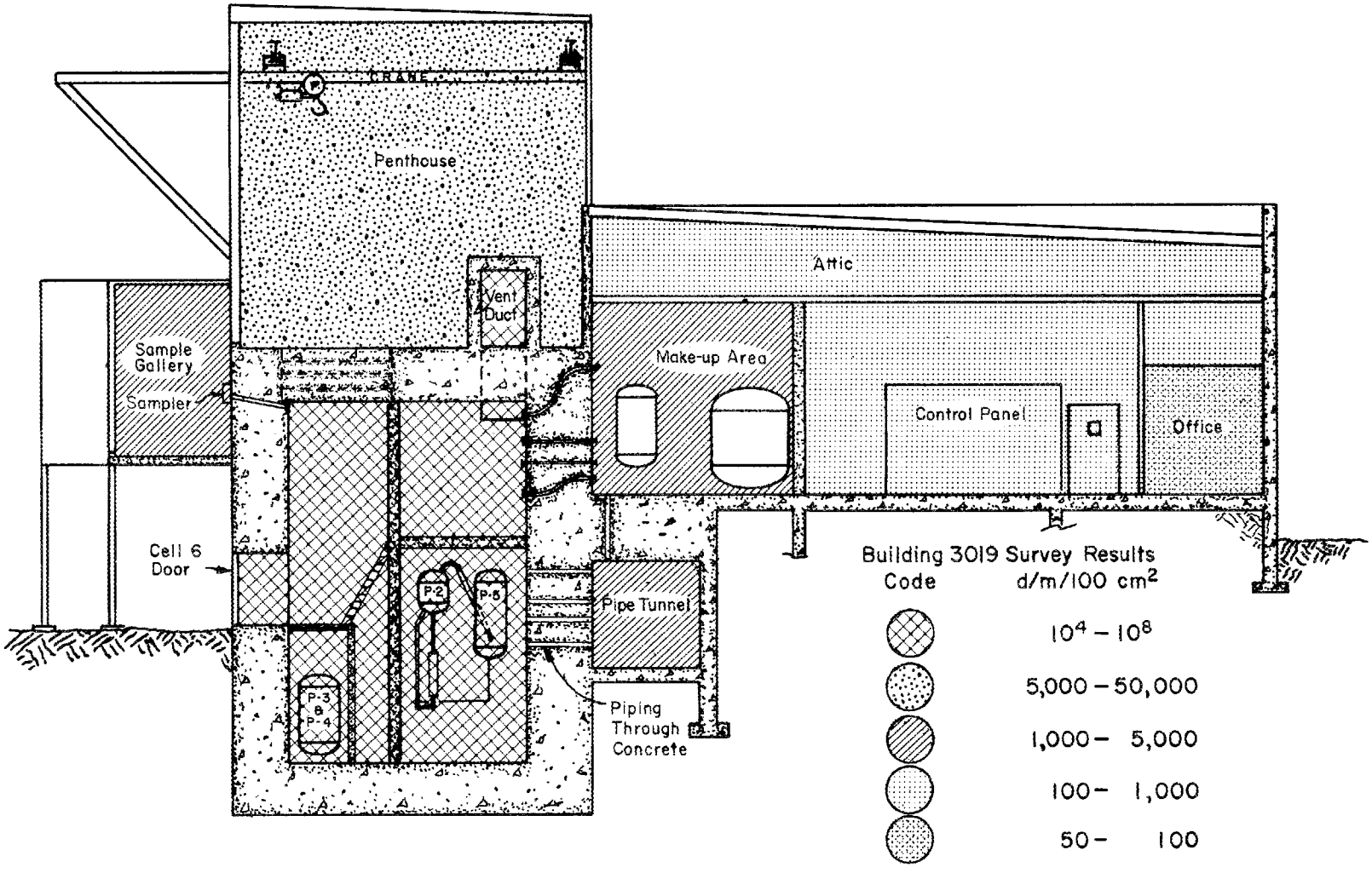

Fig. 3.4. Sectional elevation through cell 6, Radiochemical Processing Pilot Plant, showing inside contamination levels after explosion.

make right angles with the entrances. There are two entrances to the cell 6 and 7 area. One is similar to the entrances to cells 1 through 5 and is located in cell 7. The other is at grade level and is located in cell 6.

A sampling gallery running the entire length of the process cells is located on the south side of the building near the top of the cells. Radioactive samples of process solutions are collected in bottles inside a shielded sample conveyor by a recirculating air-lift sampling system.

On top of the cells is the penthouse, where are located a bridge crane for moving equipment into and out of the cells, instrument transmitter racks, and miscellaneous pieces of process equipment which are either nonradioactive or are unit-shielded.

Nonradioactive process solutions are prepared in the cold solution makeup area, located just outside the north wall of the cells.

Next to the makeup area is the control room area and next to this, along the north side of the building, are offices.

The area designated the pipe tunnel is located under the makeup area and is intended for motor-driven equipment, such as column pulsers, which requires regular or frequent maintenance and is connected directly to highly radioactive process equipment. The east end of the pipe tunnel is a storage area where uranium-233 is kept in critically safe tanks.

At the time of the explosion, processing cells 1 and 2 and the east portion of the sample gallery, penthouse, makeup area, control area, and offices were being used for the pilot plant demonstration of the volatility process for recovering uranium from fused salt reactor fuels. Cells 3 through 7 and the rest of the offices and operating areas contained the first codecontamination cycle of the Power Reactor Fuel Processing (PRFP) pilot plant. The evaporator that exploded was located in cell 6 directly opposite the cell 6 door.

3.2 P-15 Evaporator Loop

The evaporator that exploded was one of two intercycle evaporators used to concentrate aqueous uranium-plutonium strip column product. This evaporator (Fig. 3.5) was not a single piece of equipment but consisted of a steam stripper (P-5), a vapor separator (P-2), a 25-sq ft heat exchanger (P-7), and connecting piping. Overhead vapor was condensed in a 35-sq ft heat exchanger (P-8) and was collected in a condensate catch tank (P-6)

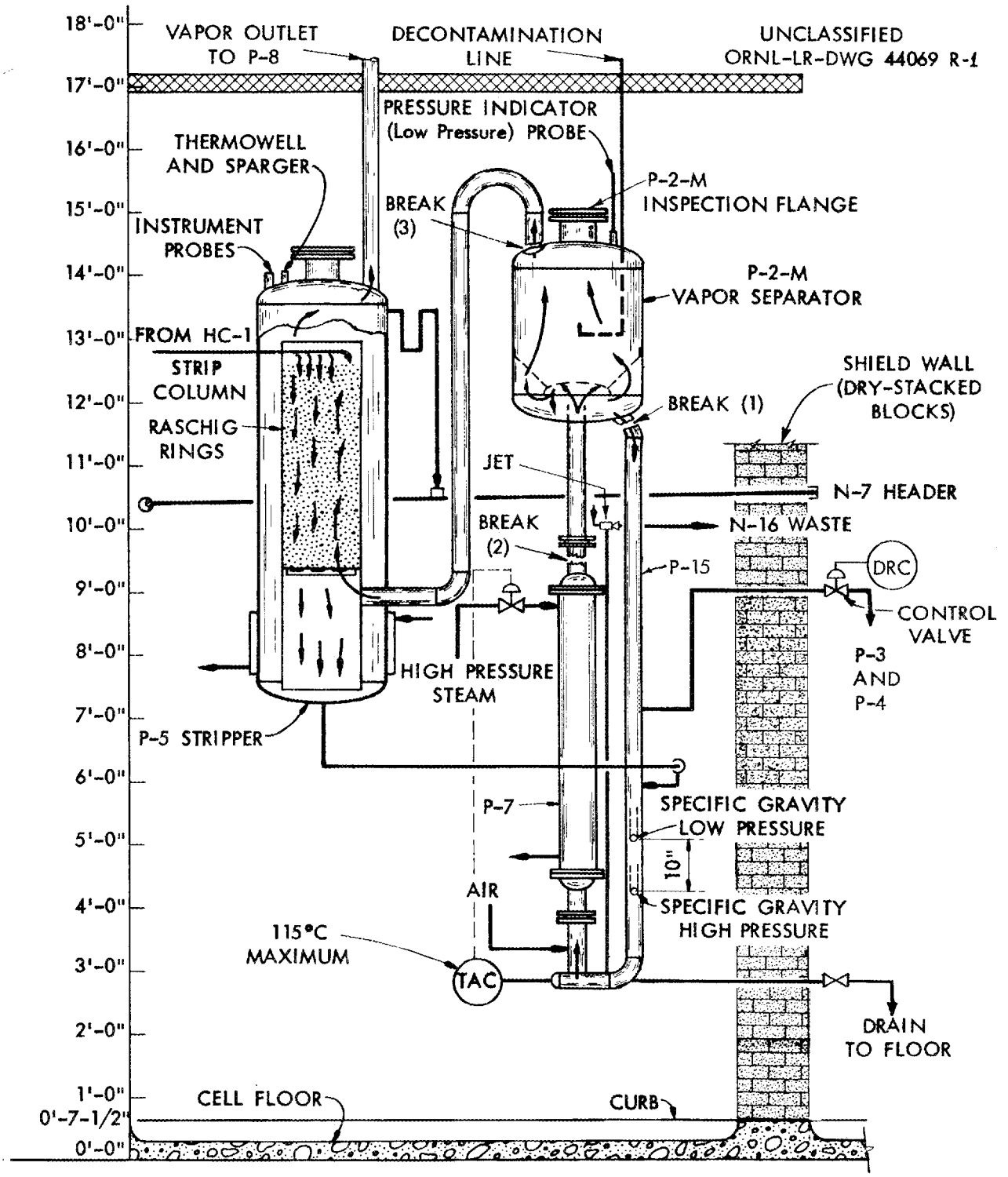

Fig. 3.5. Intercycle evaporator.

which was shared by the two evaporator loops. In normal operation, the uranium-plutonium-bearing aqueous stream from the strip column flowed by gravity through the steam stripper (for removal of dissolved or entrained organic) to the natural-convection evaporator loop. Steam and entrained liquid rose in the heated section into the vapor separator, from which the liquid returned to the bottom of the evaporator for another pass. The vapor was routed through the packed section of stripper P-5, where it served as the stripping medium, and then to the overhead condensate collection system.

The valve in the drain line was manually operated, and its use required that personnel enter the processing cell. The product outlet valve was remotely operated. Therefore it was usually used for draining the evaporator even though the product outlet line was located on the side of the natural convection loop such that 16.5 liters of solution remained in the evaporator.

3.3 Evaporator Decontamination

On November 18, 1959, decontamination of the P-15 evaporator was started. The subcell containing the P-15 evaporator contained radiation levels above 100 r/hr (hard-shell cutie pie) and caused excessive background levels throughout cells 6 and 7 and in the area south of Bldg. 3019 (through cell 6 door). Decontamination was required to allow useful working times for equipment maintenance and for installation of more shielding around the evaporator subcell.

The evaporator was treated with Turco Decon 4501,[1] a proprietary decontaminating agent which was heated 2 hr, and the evaporator was drained to the 16.5-liter residue. Fifty gallons of water was flushed through the evaporator. Then 30% nitric acid was introduced, heated without volume reduction 45 min, and drained to the 16.5-liter heel. This step was followed by a water rinse.

Radiation levels had not decreased. Therefore the manually operated drain valve was opened and the evaporator was flushed with 200 gal of water, 400 liters of hot 25% sodium hydroxide, and 300 gal of water. The manually operated drain valve was closed and the evaporator was empty except, possibly, for liquids that might be clinging to the walls.

Still radiation levels had not been measurably reduced, and it was decided to make another series of Turco Decon 4501 and nitric acid treatments. Although the intention was to follow the usual procedure of making a Turco Decon 4501 treatment, flushing with water and then making a nitric acid treatment, the instructions to shift personnel were written to "make Turco, acid, and water" treatments of the evaporator. The shift foreman knew that this was not the normal procedure but, thinking that the water flush had been omitted on other occasions, did not question the written sequence.

Turco Decon 4501 was heated in P-6 by recirculating with the internal steam jet and was added to the evaporator and heated 2 hr during which time the volume was decreased about 10% by boil-off and leakage through the product outlet valve. Because the Turco Decon 4501 had been diluted by steam condensation during heating and transfer by steam jets, it was more dilute than as received. The evaporator was drained through the product outlet line, leaving 16.5 liters.

About 270 liters of 4 N nitric acid was added to the evaporator, and the evaporator heat was turned on. Usually this acid was heated to 85°C, but this time it was boiled in the hope that the vapor spaces could be subjected to the acid treatment.

The evaporator exploded after 85 min of boildown. About 205 liters had been boiled off to the condensate catch tank and 35 liters had leaked through the product outlet valve to the product catch tank, leaving only the equivalent of 48 liters of concentrated solution in the evaporator when it exploded.

3.4 Events Following the Explosion

The three chemical operators and foreman, who were in the control room of the Power Reactor Fuel Processing (PRFP) Pilot Plant, heard the explosion and felt the building shake. They looked into the solution makeup room, which is adjacent to the control room, and saw that it was filling with a fog. Four people from the analytical chemistry laboratory, two operators from the Volatility Pilot Plant, and two of the PRFP operators left the building. The foreman actuated the evacuation alarm and called guard headquarters to request assistance from a Health Physics surveyor. He and one operator donned gas masks and looked into the solution makeup

room again. They noted no activity in the control room (checked with a cutie pie) and found nothing significant in the solution makeup room. Then, less than 2 min after the explosion, they evacuated the building. The foreman told the Laboratory shift supervisor that an explosion had occurred in cell 6 and, while the Laboratory shift supervisor issued calls for assistance from various members of the Laboratory shift organization, the foreman and the operator returned to the building momentarily to shut off water and steam and to secure operations.

Meanwhile, a Health Physics surveyor who was walking from the Oak Ridge Research Reactor (Bldg. 3042) to the Graphite Reactor (Bldg. 3001) heard the explosion, returned to his truck which was near Bldg. 3042, turned on his instruments, and drove toward Bldg. 3019. When opposite cell 6, he noticed that the cell door was open and a concrete block wall which had been in front of the door was knocked down. He surveyed the street with a Geiger counter, finding up to 15 mr/hr β-γ activity, and took smears to be counted later. ("Taking a smear" involves wiping a piece of filter paper over a surface. When the filter paper is monitored for radioactivity, or "counted," the amount of radioactivity transferred from the surface to the filter paper is determined.) The cell door was located inside a security fence and could not be approached from the street side of the building. The surveyor proceeded to the west gate of the 3019 security area, where he surveyed the evacuees for β-γ activity and found none. It was then about 5 min after the explosion.

Operating, Health Physics, and management personnel were called to the Laboratory and access to the affected area was restricted. A plan was initiated to evaluate the incident and start cleanup operations. Security guards at the Laboratory gates were instructed that only persons required at the scene of the explosion should be allowed to enter.

The evacuees were surveyed for α contamination and none was found.

The foreman and a supervisor put on gas masks and looked into cell 6 through the door that had been blown open. The light was poor, but they could see that the unmortared block wall in front of the evaporator subcell had been knocked over and equipment in the subcell disarranged.

At midnight the Laboratory gates were opened to allow the shifts to change and those leaving were surveyed for contamination. Shortly after

midnight, α activity was reported on a filter paper that had been removed from one of the Bldg. 3019 air monitors and, concurrently, a Health Physics surveyor found α contamination on his shoes. Alpha activity was then found in the street in front of the cell 6 door and on the cheeks of the supervisor who had looked into cell 6.

Laboratory waste streams were found to contain 25 to 150 α c/m/ml. Process waste was directed to a basin where it could be stored, and process water saving measures were instituted throughout the Laboratory to minimize the waste storage problem.

The Graphite Reactor Building (3001) was found to be contaminated with α activity. The reactor was shut down and the building evacuated.

3.5 Extent of Contamination

Radioactivity was released to the processing cell, to the Chemical Technology Division pilot plant operating and office areas, and to buildings and grounds in the vicinity of the Radiochemical Processing Pilot Plant as a result of the explosion. Fallout of the radioactivity was evidently rapid, and the contaminated area was only a small fraction of the Laboratory area. Alpha-emitting plutonium was the major contaminant and β-γ emitting zirconium-95 and niobium-95 were of secondary importance. The potential hazard presented by the plutonium and the cleanup effort required to contain or remove the plutonium α activity were so great as to overshadow the consequences of the zirconium-niobium contamination.

3.5.1 Methods Used to Define Contaminated Areas

Standard Health Physics techniques[2] were used to define contaminated areas and to indicate the degree to which these areas were contaminated. Gas-purged proportional counters were used for direct reading α measurements, cutie pies and Geiger counters for direct reading β-γ measurements, and filter paper smear techniques for determining both α and β-γ transferable radioactivity.

Data from Health Physics surveys were used to define contamination zones and to estimate roughly the amount of plutonium dispersed by the explosion, although it was realized that the value of Health Physics survey data for this latter purpose was questionable. The purpose of a Health Physics survey is to determine whether the area surveyed presents a health hazard

to personnel, and the absence of such hazard is indicated when a survey, made according to prescribed monitoring techniques, yields numerical results that are below established tolerances for those techniques. When an area is grossly contaminated, the fact that the contamination exists is the end result of the survey. Instruments are moved over the area faster than normally, and filter paper smears are not rubbed as hard or as long as normal. Only when contamination levels are low, because of cleanup or because the area being surveyed is only slightly contaminated, is extreme care taken to assure that the survey is being made in accordance with prescribed procedures. Calculations based on Health Physics survey data, which are intended to determine the amount of a particular isotope spread over a surface, involve estimating the relation between the amount of the isotope on the surface and the effectiveness of the monitoring technique used. In the following sections, the figures that show contamination levels in various areas are merely a convenient way of presenting Health Physics survey data. Calculations of the amount of plutonium dispersed by the explosion are, at best, order-of-magnitude estimates.

3.5.2 Radioactivity Released to Cells 6 and 7

Approximately 150 g of plutonium has been flushed from surfaces in cells 6 and 7. Most of this came from the ruptured evaporator and it has been estimated that only about 15 g was released from the evaporator subcell.

Processing cells 6 and 7 (excluding the evaporator subcell) were hurriedly smeared with paper towels. Five smears, each covering about 4 sq ft, were taken. Using the results of these smears as indicators of average contamination over the various cell surfaces, and assuming a transfer to the towel of 10% of the activity present, gave a total of 14.7 g of plutonium released from the evaporator subcell to the general cell 6 and 7 area.

3.5.3 Radioactivity Released from the Processing Cells

From the time Bldg. 3019 was built in 1943 until the evaporator exploded, controlled air flow from the occupied, low-activity areas through unoccupied, high-activity areas to an off-gas treatment facility and dis-

charge stack was the means used to limit the spread of air-borne radioactivity. The pressure rise that must have accompanied the evaporator explosion reversed the air flow momentarily, causing air-borne activity to be transported from the processing cells to other areas in the building and to the outside of the building.

Fallout of the dispersed activity was rapid, and only a small fraction of the Laboratory area became contaminated. An indication of the rapidity of fallout is the fact that the outside of a truck driven through the fallout zone by a Health Physics surveyor became highly contaminated, but the surveyor, who got out of the truck and looked through the building security fence at the cell 6 door, merely got his shoes contaminated.

a. Contamination in Bldg. 3019. Plutonium was forced from the processing cells via access holes penetrating the cell walls and was spread throughout the portion of Bldg. 3019 used as offices and operating areas by the Pilot Plant Section of the Chemical Technology Division. The walls of cells 6 and 7 contain 38 access holes, which were either completely open or partially filled with piping and which total over 6 sq ft in area. Figure 3.4 shows the contamination levels in various parts of the building. Most smears taken in the penthouse, the most highly contaminated building area outside the cells, indicated transferable α contamination of from 5000 to 50,000 d/m/100 sq cm, and direct probe measurements were as high as 800,000 d/m/100 sq cm. Most smears taken in the sample gallery, nonradioactive solution makeup area, and pipe tunnel ranged from 1000 to 5000 d/m/100 sq cm. The average α contamination in the pilot plant offices was about 50 d/m/100 sq cm. A crude estimate based on smear data indicated that ~70 mg of plutonium had been spread in the building.

The parts of the building that contain the analytical chemistry laboratories and the basement area under the analytical chemistry laboratories were not contaminated as a result of the accident.

b. Contamination Outside of Bldg. 3019. The cell 6 door, which is at ground level and opened directly to the outside of Bldg. 3019, was thrown open by the explosion, and it knocked down a dry-stacked concrete block shielding wall just outside the door. Plutonium released through this doorway contaminated nearby streets and building surfaces. The Graphite Reactor Building (3001) became contaminated by plutonium released through

the cell 6 door and subsequently drawn into and through the reactor building by the ventilation system. The affected area is indicated on Fig. 3.1, and Fig. 3.6, an enlargement of this area, shows the degree to which the area was contaminated. A calculation similar to the one used to determine that ~70 mg of plutonium was released to Bldg. 3019 indicated that ~600 mg was released from the building.

3.5.4 Cell Ventilation Filtering System

Filters in the ventilation system for the radiochemical processing cells contained about 1.5 g of plutonium, most of which was probably released to the cell ventilation system by the explosion. The concentration of the plutonium at the front of the filter bank indicated that the plutonium was retained with a high degree of efficiency.

The cell ventilation filtering system (Fig. 3.7) comprises three parallel filter banks, each of which contains seven compartments for roughing filters which are operated in parallel and two compartments for absolute filters, also operated in parallel. Each roughing filter compartment contains five deep-bed pocket filters which are in parallel and are composed of a 1-in.-thick front (first to be exposed to air flow) layer of FG-25 filterdown, followed by a 1-in.-thick layer of FG-50 filterdown. Each absolute filter compartment contains three parallel filters. Thus cell ventilation air flow from the cells passes through one layer of FG-25 filterdown and one layer of FG-50 filterdown in a roughing filter and then through an absolute filter before being discharged to the stack. The roughing filters are 93.9% efficient, by particle count, on atmospheric dust retention, and the absolute filters, which are either asbestos or glass fiber, are 99.95% efficient on retention of 0.3-μ size particles of dioctyl phthalate.

Examination of three large sections of roughing filter and four sections of absolute filter indicates that 1.5 g of plutonium, of the same isotopic concentration as the plutonium found in the evaporator, and 75.8 g of uranium were collected by the system. Principal γ activities were zirconium, niobium, and ruthenium. The roughing filters contained 98.8% of the plutonium, 97.8% of the γ activity, and 88.3% of the uranium found in the filtering system. It is probable that no alpha activity penetrated to the back of the absolute filters.

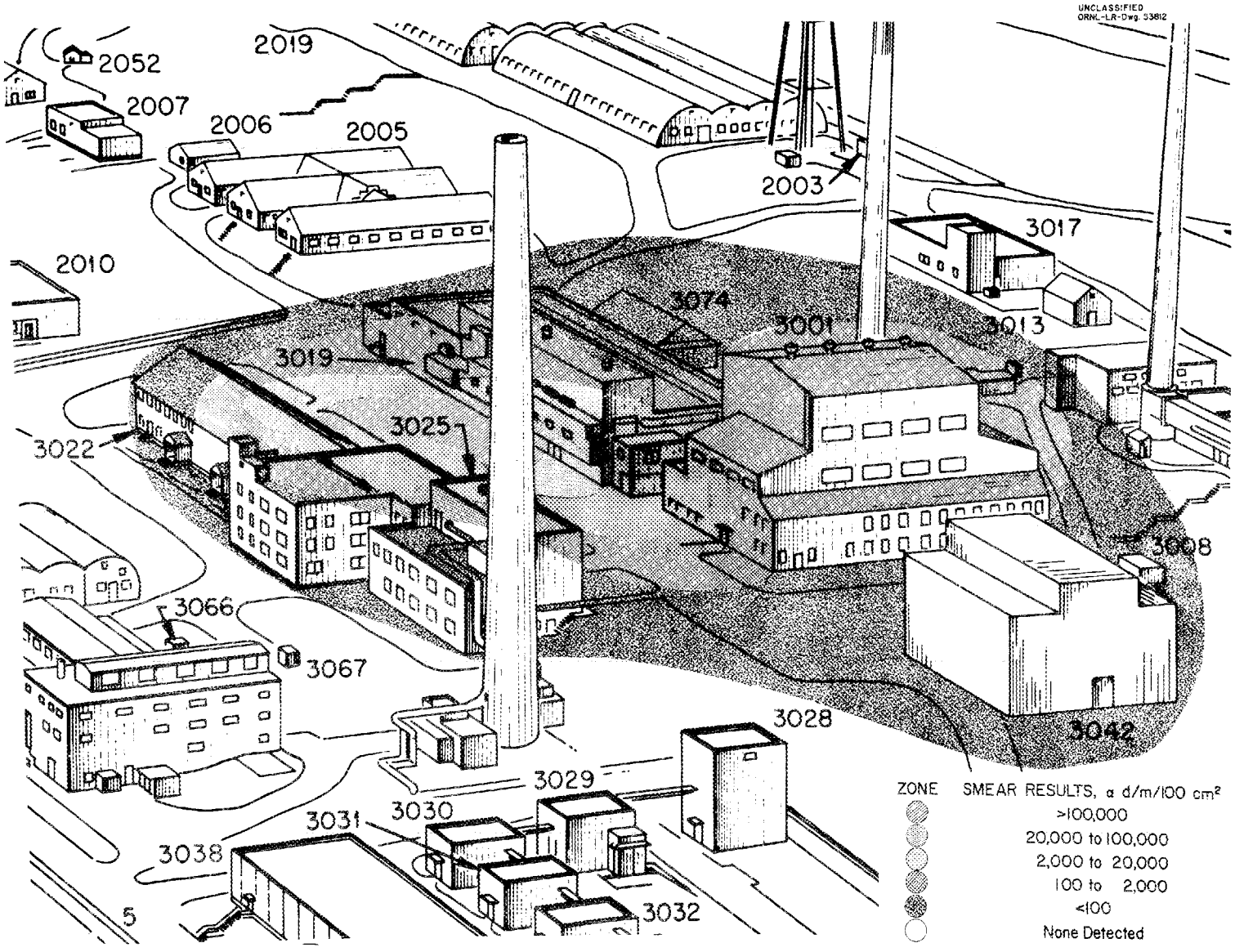

Fig. 3.6. Plutonium fallout zones after explosion.

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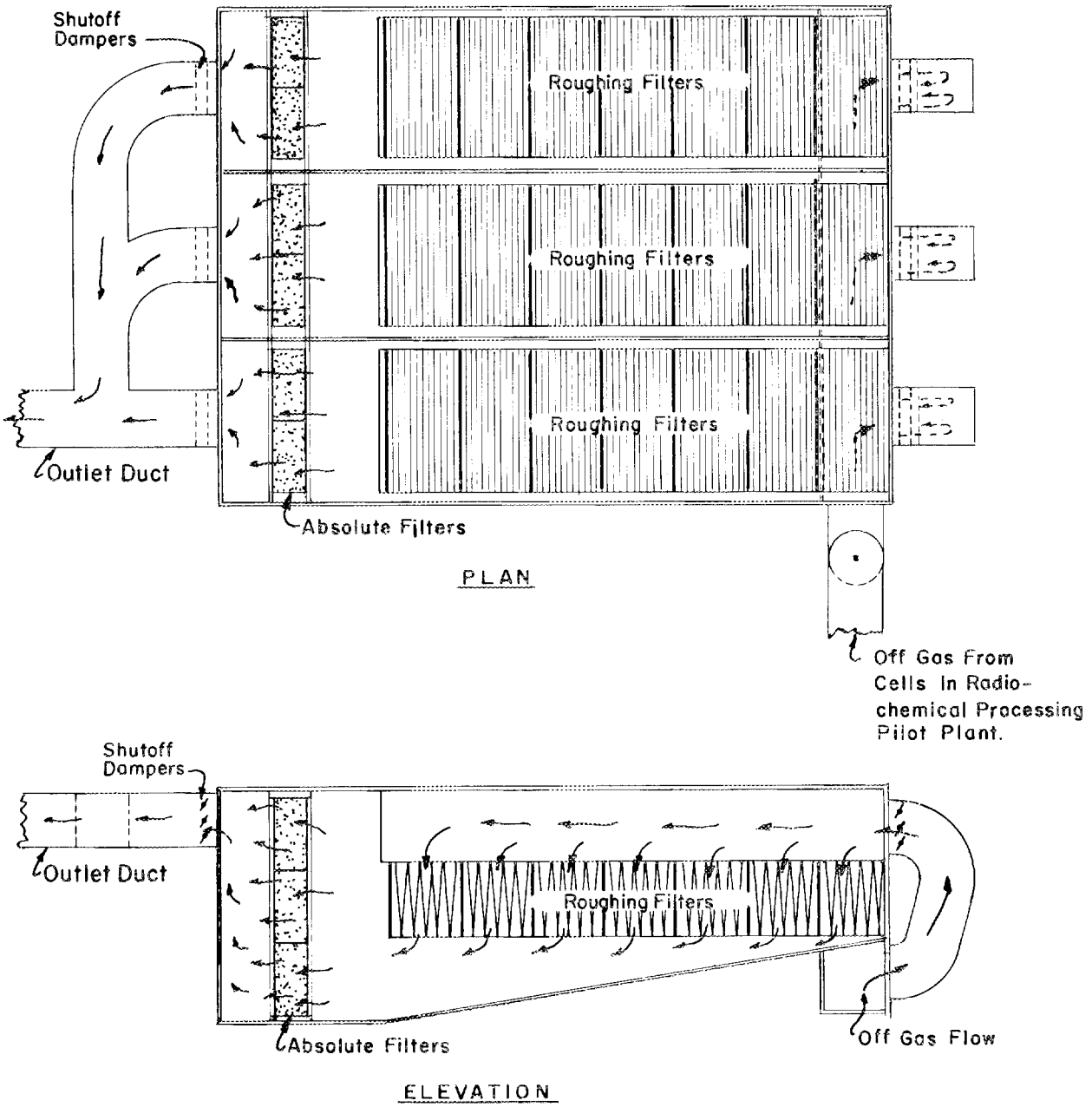

Fig. 3.7. Radiochemical Processing Pilot Plant cell off-gas filter facility.

3.6 Blast Damage to Equipment by the Explosion

The explosion was a minor chemical explosion and physical damage was confined to the equipment in the evaporator subcell, a few pipes which were connected to this equipment, and the unmortared concrete block shielding walls which were in front of the subcell (Fig. 3.8) and in front of the cell 6 door. Access to the cell area has been limited, and it has not been possible to make a thorough examination of the ruptured evaporator. However, photographs of the subcell plus a few observations of the equipment have made possible a reasonably accurate description of the damage (Fig. 3.9).

The vapor separator was torn apart by the incident. A large, badly deformed section that originally was part of the side and top and bottom was found lying on the floor in the subcell (Fig. 3.10). The remainder of the separator was found wrapped around the steam stripper which was dented by the impact (Fig. 3.11).

The convection loop was completely separated from the vapor separator as the result of mechanical failures in the heat-affected zones surrounding the welds that connected the two ends of the convection loop and the vapor separator. The flared end of the heat exchanger nozzle, which was flanged to a 2-in.-dia pipe, which, in turn, was welded to the bottom of the vapor separator, failed circumferentially and the 2-in.-dia pipe was found lying on the floor of the subcell. The bottom flange on the heat exchanger was found to be open, and apparently the flange bolts or nuts were stripped.

Water supplied to the heat exchanger jacket after the explosion leaked into the convection loop and, in addition to leaking from the damaged bottom flange, ran out the convection loop cold leg although the top of this cold leg is at a higher elevation than the top of the heat exchanger leg. The tube bundle appears to be badly damaged.

Miscellaneous pipes which are or were connected to the evaporator are bent or twisted but they do not seem to be damaged so badly as to prevent flow through them.

The shielding walls knocked down as a result of the explosion were constructed of barytes concrete blocks, which were dry-stacked. Very little force was required to bring about their collapse. Some of the blocks were

Fig. 3.8. Debris in front of evaporator subcell after explosion.

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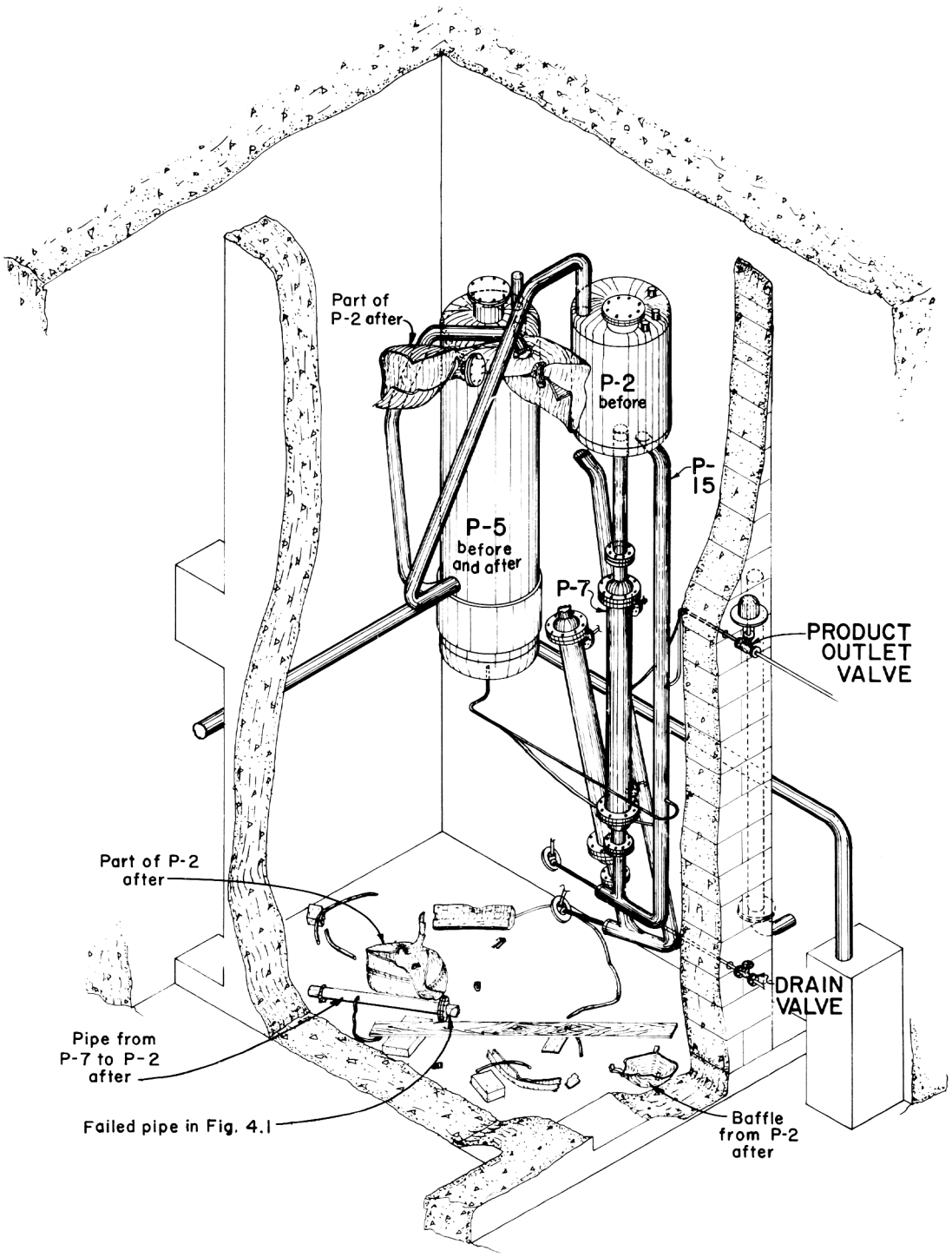

Fig. 3.9. Intercycle evaporator before and after explosion.

Fig. 3.10. Floor of evaporator subcell after explosion.

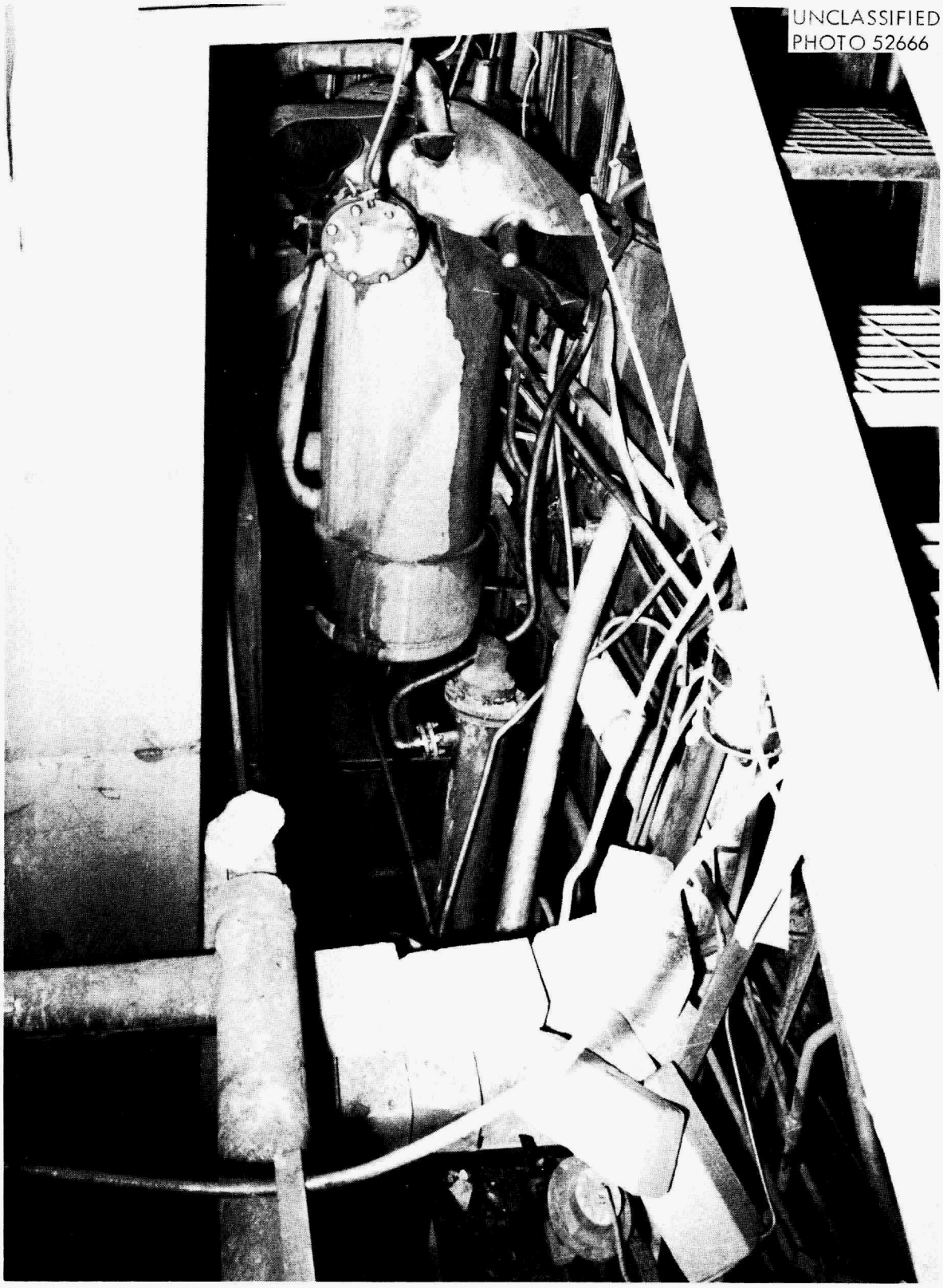

Fig. 3.11. Upper portion of evaporator subcell after explosion.

chipped slightly but, in general, individual blocks received little physical damage.

4.0 ANALYSIS OF THE EXPLOSION

4.1 Possible Nuclear Origin

The explosion and subsequent release of radioactivity were not caused by a nuclear excursion, but at the time of the incident there existed reason to suspect that a nuclear excursion, involving either uranium-233 or plutonium-239, had occurred. Uranium-233 is stored in geometrically safe storage tanks in the east end of the pipe tunnel and, at the time of the explosion, these tanks contained about 40 kg of this fissionable isotope.

During the chemical processing programs entitled the "Plutonium 240" programs, which were conducted from August through November, 1959, the determination of a plutonium material balance was encumbered by problems arising from process solution characteristics, sampling, and method of analysis. The fuel solutions contained a larger fraction of solids than previously experienced. Because of the high burnup of the feed metal, air sparging of the feed solution before sampling had been held to a minimum to limit the amount of radioactivity discharged to the vessel off-gas system. The analytical method used originally (and which previously had been successful in providing a plutonium material balance) was to centrifuge the sample and analyze the supernatant liquid by TTA (thenoyltrifluoroacetone) extraction method.[3] It has been determined that this analytical method was not useful for these samples because the solids contained much of the plutonium, and further, this plutonium was in a nonextractable form. The result was that on Nov. 20, 1959, a plutonium balance was not complete; the possibility existed that more than a critical mass could not be accounted for. It was shown later that the material balance for the 27 kg of plutonium processed during the programs was minus 4,570 $\pm$ 3,250 g. Approximately 1500 g of plutonium could not be accounted for by errors in the balance.

However, within 3 hr after the explosion the possibility that the explosion had been nuclear in origin was considered remote. Threshold flux detectors distributed throughout the building were examined and no neutron activation was detected. These detectors were collected by the Health Physics Division area leader for the building, and he noted that there had been no increase in radiation background levels in the building as a result of the explosion. Further, none of the smears or air samples taken immediately after the explosion contained any short-lived fission products.

4.2 Chemistry of the Incident[4]

The intercycle evaporator explosion probably was caused by rapid reaction of the products of the extensive nitration by concentrated nitric acid of the proprietary decontaminating agent Turco-4501. This is a strongly basic solution of the alkali salts of various organic hydroxy acids, amines, surface active agents, and phenol. The products of nitration are potassium nitrate (a component of black gun powder), nitrate salts of the amines, nitrate esters, and picric acid. Potassium nitrate crystals were found in the evaporator after the explosion. These compounds, especially picric acid, are thermodynamically unstable and are readily ignited.

It is possible that tributyl phosphate (TBP) and its degradation products di- and monobutyl phosphoric acids could have been flushed from the steam stripper to the convection loop and that reactions of these materials with hot nitric acid may have been contributory to the explosion; however, there is no evidence to suggest the presence of significant quantities of TBP or its degradation products in the evaporator at the time of the explosion.

4.2.1 Chemicals Inventory Preceding the Explosion

The manually operated drain valve was closed at 3:00 p. m. on Nov. 20, 1959, and, except that tributyl phosphate degradation products may have been adhering to the metal surfaces, the history of operations prior to this time had no effect on the subsequent explosion. A solution of 200 liters of Turco Decon-4501 and 62 liters of water (steam jet dilution) was added to the evaporator. The lower 15% of the Raschig ring container in the steam stripper was immersed in the solution at the start of a 2-hr boildown, dur-

ing which the solution was concentrated to 238 liters. The solution was drained through the density control valve to product receiver P-3, leaving the drain heel of 16.5 liters (13.9 liters of Turco Decon 4501 and 2.6 liters of water).

The next addition to the evaporator loop was 270 liters of 4 M nitric acid, which was added through the vapor separator in such a way that it flowed into the cooler leg of the convection loop, displaced about 1.6 liters of the Turco-water mixture, and then flowed into the steam stripper. The total volume of liquid in the system was 286 liters, of which 29 liters was in the convection loop of the evaporator; this 29 liters was composed of about 12.6 liters of "as received" Turco Decon 4501 and 16.4 liters of 4 M nitric acid.

From potentiometric titration by strong acid the following approximate composition of Turco Decon 4501 was deduced:

Strong base	3.9 N
Weak organic acids	1.7 N
Stronger acids, such as phenol	3.1 N

It is estimated that prior to starting the last boildown, there was in the heat exchanger loop only about 5.2 moles of nitric acid per liter of Turco Decon 4501. Thus, at the start of boiling the mixture in the loop was probably still alkaline. It is probable that some products of the degradation of tributyl phosphate (TBP) were also in the intercycle evaporator. Di- and monobutyl phosphoric acid are known to form with some cations, e. g., Fe(III), Zr, and U(VI), compounds that are only slightly soluble in aqueous solutions such as were normally processed in the P-15 evaporator. Also, an appreciable fraction of tributyl phosphate dissolved or entrained in these solutions would be decomposed in the steam stripper and in the convection loop. The inventory of these materials at the time of the last addition of the decontaminating agent is unknown.

During the 85-min interval between the start of boiling and the explosion, 205 liters of 1.42 N acid was distilled into the condensate catch tank and 35 liters of solution leaked through the product outlet valve to product catch tank P-3. Thus, the average rate of flow of 4 M nitric acid from the steam stripper to the convection loop was about 2.8 liters/min.

At this rate about 5 min would have elapsed from the start of boiling to the neutralization of the Turco Decon 4501. Conversion of the amines to substituted ammonium nitrates and nitration of the organic material would then begin.

Just prior to the explosion, the evaporation system probably had the following chemical inventory: (1) in the steam stripper P-5, approximately 30 liters of 30-35% nitric acid (5.6 - 6.7 M); (2) in the convection loop—heat exchanger combination, approximately 18 liters of the products from extensive nitration of the Turco Decon 4501 and some unknown quantity of phosphoric acid and butyl nitrate obtained from the nitration of TBP degradation products. The convection loop—heat exchanger section may have contained solid alkali nitrate and possibly 2-3 kg of picric acid.

4.3 Mechanics of Evaporator Failure

In investigating an explosion that has occurred in a piping-pressure vessel system and attempting to classify the type, one normally starts by considering the location and position of fragments and the distortion of tube sheets, baffles, and other parts of the flow path. The high α contamination level and the initially high levels of penetrating radiation in cell 6 made it difficult to locate and/or examine all fragments. Conclusions were based on photographs of the area (Figs. 3.8, 3.10, and 3.11), reports from a few people who had entered the cell, visual inspection of the area from a distance of about 20 ft, and a complete metallographic examination of only one fragment.

4.3.1 Metallographic Examination of Failed Pipe

Figure 4.1 is a photograph of a piece of a flared pipe which connected the convection loop heat exchanger to the long nozzle that was welded to the bottom of vapor separator P-2. In Fig. 3.5 the piece is shown between "Break (2)" and the flanged connection just above "Break (2)." It can also be seen protruding from the flange that is near the bottom in the center of Fig. 3.10. (See also Fig. 3.9).

Metallographic specimens were cut from the two areas of interest (Fig. 4.1) with a hacksaw. The cuts were made parallel to the length of the pipe.

The specimens were mounted in thermosetting plastic and hand ground on 240 through 600 grit silicon carbide paper to the depth of interest.

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Fig. 4.1. Failed pipe from ruptured evaporator.

They were then polished on microcloth with alumina, 1-μ diamond, and Linde B, in that order. Carbon tetrachloride served as the lubricant for all grinding and polishing operations. Specimens were then swab-etched with a ferric chloride solution and rinsed with carbon tetrachloride.

The normal structure of the pipe away from the areas of fracture is shown in Fig. 4.2a. The structure is equiaxed, with annealing twins being seen in some of the grains and can be considered typical of stainless steel. The wavy striated background structure, which appears in this photomicrograph faintly and is seen to varying degree in the remaining photomicrographs, is parallel to the direction of working. Coring-type segregation occurs during casting of the stainless steel, which produces variations in composition on a microscale. These regions of segregation are elongated during fabrication and are shown as a result of staining during etching.

The structure in Area 1 (Fig. 4.1) is shown in Fig. 4.2b. The grains appear equiaxed with no evidence of plastic strain in this area. The structure in Area 2 (Fig. 4.1), seen in Fig. 4.2c, on the other hand shows definite grain distortion and numerous fine mechanical twins. As one moves away from the immediate fracture area (Fig. 4.2d), the grain distortion is no longer evident, although mechanical twinning is still quite pronounced.

On the basis of these observations, it can be concluded that plastic deformation at the one side of the pipe preceded failure. Furthermore, it appears that once failure occurred in this area, the crack that resulted propagated rapidly through the remaining material without further plastic deformation.

4.3.2 Conclusions

1. The explosion occurred in the bottom of vapor separator P-2 or in the warm leg of the evaporator loop. The way in which the baffle that was in the vapor separator was deformed indicates that the force came from beneath it (Fig. 4.3).

2. The peak explosion pressure was probably in excess of 1100 psi since that is the estimated rupture pressure of vapor separator P-2.

3. The rate of pressure rise was such that the vapor outlet from the top of stripper P-5 could not handle the gases generated. Further, the pressure increased sufficiently fast in P-2 to rupture it before the pres-

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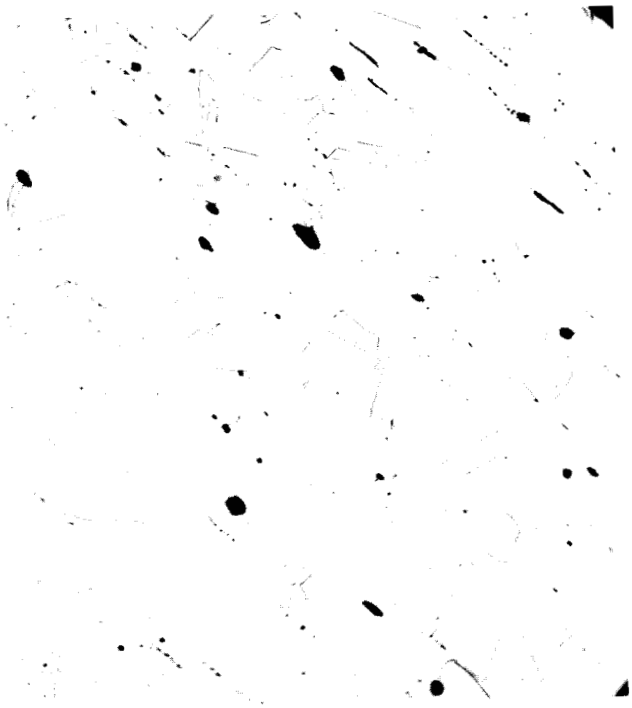

Fig. 4.2a. Normal structure

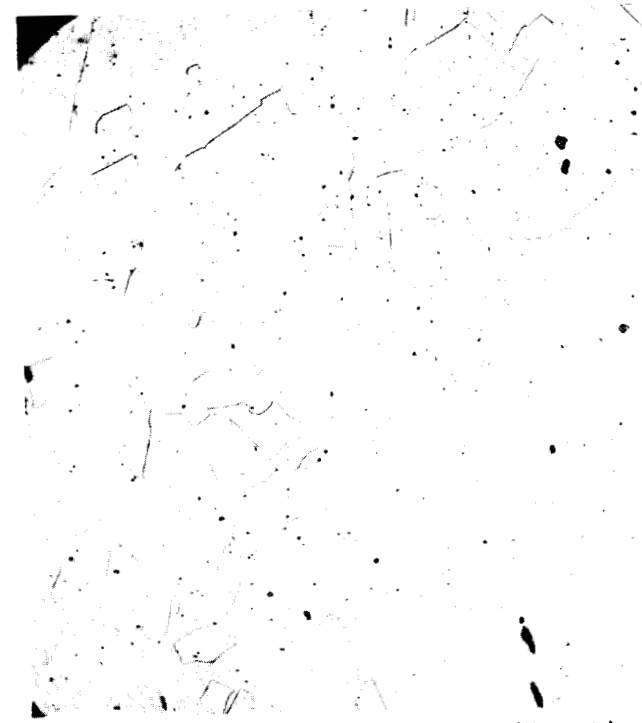

Fig. 4.2b. Area next to fracture; area 1 (Fig. 4.1.). No evidence of plastic deformation.

Fig. 4.2c. Area next to fracture; area 2 (Fig. 4.1). Mechanical twinning and grain distortion apparent.

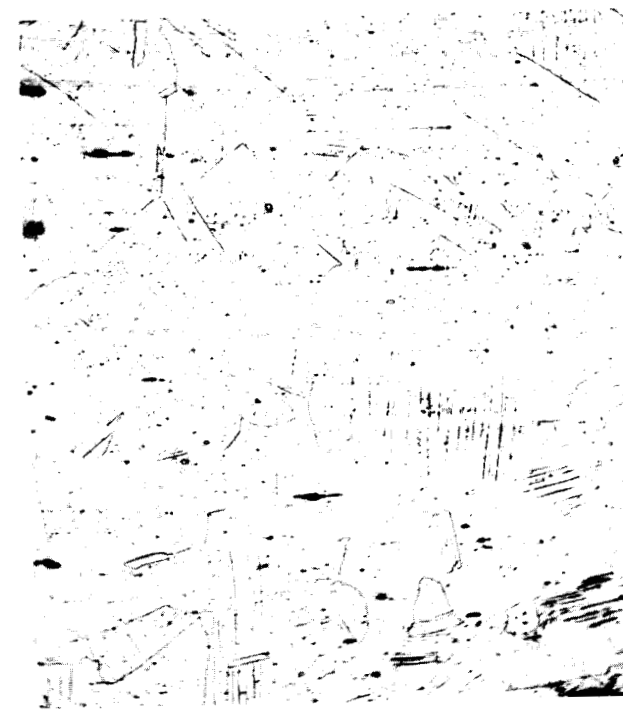

Fig. 4.2d. Slightly further away from the fracture than area shown in Fig. 4.2c; area 2 (Fig. 4.1). Mechanical twinning present.

Fig. 4.2. Photomicrographs (125 x) of specimens cut from failed pipe from ruptured evaporator.

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Fig. 4.3. Baffle from vapor separator P-2.

sure could increase in the P-5 stripper to visibly distort it.

4. Since the elbows in the piping to and from the vapor separator P-2 are not cracked, or even visibly distorted, it is unlikely that a shock wave traveled through the lines. This opinion appears to be supported by the fact that the bolts in the piping flange below heat exchanger P-7 are visibly elongated on one side, such as might occur from a piping reaction produced by the explosion instead of from a shock wave.

5. There was no shock wave in the warm leg of the convection loop. Metallographic examination of specimens cut from the failed pipe (Fig. 4.1) which connected the convection loop heat exchanger to the vapor separator indicates that plastic deformation on one side of the pipe preceded failure.

5.0 CLEANUP AND DECONTAMINATION

The objectives of the program for cleanup and decontamination were (1) immediate containment of the radioactivity to prevent its spreading to other Laboratory areas or from the Laboratory, (2) decontamination of areas or buildings that were slightly contaminated and could easily be made available for service, (3) decontamination of streets and exterior surfaces of buildings, and (4) decontamination of the Graphite Reactor Building (3001) and the Radiochemical Processing Pilot Plant Building (3019).

An extensive program of checking Laboratory areas and personnel was carried out for about 3 weeks after the activity release. Occasionally people were found to have contaminated shoes, and building areas were sometimes found to be slightly contaminated, but these occurrences were not well documented. The checking was maintained until α activity was no longer discovered in the ORNL area, indicating that the radioactivity had been completely contained.

5.1 Emergency Measures

Containment of the radioactivity was the first consideration. A gross washdown of the area was not attempted: first, because contamination might be washed into crevices where it would be inaccessible for cleanup but would be present as a contributor to high air-activity levels; and second, because the amount of water that could be held in the waste storage basins was limited. It was decided to fix the activity until it could be removed in an orderly way at some later time. Badly contaminated roads and the roof of Bldg. 3025 were resurfaced with emulsified asphalt and gravel. Paint was sprayed on the roof and north wall of Bldg. 3022, the south wall of 3019, the general area inside the 3019 security fence (including all equipment and material), barrels and miscellaneous material on the north side of 3019, the complete exterior of the 3019 sheet metal penthouse, the sidewalk on the north side of 3022, and grass on the north side of 3022 and 3025 and on the south and east sides of 3001. The interiors of Bldgs. 3022, 3025, 3042, 3010, 3012, and 3044 were cleaned as needed to make them ready for occupancy by Nov. 23, 1959.

As a precaution against the possibility that, in the haste of cleanup, some α contamination had been overlooked, Bldgs. 3074, 3022, 3025, 3005, and

3042 and the analytical chemistry laboratory in Bldg. 3019 were designated controlled areas. No food was allowed in these buildings; people using these buildings were instructed to wash their hands carefully and have them checked for radioactivity before eating and to make frequent checks for radioactivity on their hands and feet. The pilot plant portion of Bldg. 3019, Bldg. 3001, and the road south of these buildings were designated exclusion areas; only personnel actively engaged in cleanup operations were permitted there.

Except for these two buildings and this section of roadway, all Laboratory areas were back in service by Nov. 23.

5.2 Decontamination of Outdoor Areas

The exterior surfaces of the buildings in the area that was contaminated (Fig. 3.6) and the surfaces of the roads and grounds in the vicinity of Bldg. 3019 were given some sort of specialized treatment as indicated below. No pretreatment was given to any of the surfaces, and no exterior surfaces were washed before painting.

5.2.1 Building 3019 (Radiochemical Processing Pilot Plant)

The security fence south of Bldg. 3019 enclosed a temporary thorium-storage area containing a stainless steel tank, a welding machine, a sheet metal—enclosed area containing three stainless steel thorium product decay tanks and a source and special materials vault, a dump truck, an acid transfer truck, and some stainless steel pipes in addition to the permanent thorium-storage pit which contains three stainless steel tanks. All exposed surfaces of this equipment and material were painted between Nov. 21 and Nov. 23 to fix the contamination.

Later, everything except the tanks in the permanent storage pit was removed to the burial ground to await decision on its final disposition. The security fence was assumed to be contaminated and was removed. The roof above the processing facility (including the penthouse and sampling gallery) was covered with tar and chips to a depth of up to 2 in. The aluminum siding on the sampling gallery and penthouse was painted the original color, and all duct work between the building and the 3019 stack was painted. The south side of the building below the sampling gallery was painted white. Later the white paint was removed with paint remover and the wall repainted.

5.2.2 Building 3022 (Engineering and Mechanical)

The roof and north wall were sprayed with green enamel paint during the original campaign to fix the α activity. Since then, two coats of aluminum paint have been applied to the roof, and the north and west sides of the building have been painted to match the original color. Fourteen air conditioners were removed from windows on the north side and an air supply duct from the south side was installed.

5.2.3 Building 3025 (Solid State Physics)

The roof was sprayed with tar and chips and then painted, and a cooling tower and some duct work on top of the building were painted. The north wall (ordinary red brick) was wiped with detergent and water and sealed with two coats of masonry sealing compound.

5.2.4 Building 3001 (Graphite Reactor)

The Bldg. 3001 roof was coated with aluminum-fiber roofing compound to a maximum depth of 0.25 in. All four external walls of the building were repainted the original aluminum color.

5.2.5 Building 3005 (Low Intensity Test Reactor)

The Bldg. 3005 roof on the east and west wings was coated with aluminum-fiber roofing compound in the same way as Bldg. 3001. The entire building was repainted the original color.

5.2.6 Buildings 3003 (Fan House), 3004 (Demineralized Water Plant), and 3008 (Source and Special Materials Vault)

These buildings were completely repainted.

5.2.7 Roads and Grounds

The surface of Hillside Avenue between Third Street and Bldg. 3042 (approximately 645 linear feet), which had been covered with tar and chips, was removed to a depth of 1 ft and replaced with gravel. The paved area between Bldgs. 3001 and 3042 was treated similarly. When the outdoor cleanup operations were completed, these areas were paved. The roadway north of Bldg. 3019 and south of Bldg. 3074 was covered with emulsified asphalt and gravel. Ground was removed in the area enclosed by Bldgs. 3019, 3022, 3025, 3001, and 3042. Various concrete pads were ground with a terrazzo floor grinding machine.

5.3 Decontamination of the Graphite Reactor Building

The ORNL Graphite Reactor was not in operation from Nov. 20 through Dec. 22, 1959. Every laboratory and work area in the building was contaminated with plutonium drawn into and through the building by the ventilation system. About 11,000 man-hours was required to vacuum clean, wash, and survey the building. The decontamination specifications (Sect. 5.4.2) for the Radiochemical Pilot Plant were also used for the cleanup of the Graphite Reactor building.

5.4 Decontamination of the Radiochemical Processing Pilot Plant

All Chemical Technology Division areas of the Pilot Plant (the east portion of Bldg. 3019) were contaminated by plutonium as a result of the evaporator explosion. Access to these areas was limited to cleanup crews; all other personnel normally assigned to this area were relocated in other ORNL buildings.

Offices and control room areas were first to be decontaminated and were reoccupied on Jan. 14, 1960. Decontamination of processing cells 1-5 and the makeup area, sampling gallery, pipe tunnel, and penthouse area was started just prior to the end of office decontamination, but cleanup of processing cells 6 and 7 was delayed 8 months until building modifications could be made to provide for improved containment of radioactivity.

5.4.1 Contamination Levels

Highly radioactive materials have been processed in Bldg. 3019 since 1943 (Appendix B). The processing cells and adjacent operating areas have become contaminated on many occasions. Until recently, the only useful means for monitoring α contamination was to "smear" suspected areas with filter paper swipes and monitor these swipes in a laboratory α counter. Reliable and sensitive portable α survey meters did not exist during the first 10 to 15 years of operation of this facility, and α activity that was etched into various surfaces or that found its way into cracks and crevices remained undetected. Also, it was fairly common practice in earlier years to cover moderate α contamination with paint or concrete if it could not be cleaned up conveniently by washing. Although a large quantity of radioactivity was spread by the evaporator explosion, the difficulty encountered in decontaminating Bldg. 3019 was compounded by

contamination that had been spread and then covered during previous programs. In many instances the removal of paint, concrete, or metal surfaces, which almost certainly removed the contamination resulting from the explosion, uncovered contamination that had been deposited long ago and it was not unusual that contamination levels increased instead of decreasing.

5.4.2 Decontamination Specifications

Building areas were decontaminated to noncontamination zone α activity specifications, or, if cleaning to these specifications was not feasible, areas were decontaminated only to within 10 times these specifications and the surfaces were color coded with orange enamel and then covered with either paint or concrete. The specifications were:

	Direct Reading	Transferable
Max.	300 d/m/100 cm^2	30 d/m/100 cm^2
Avg.*	$\leq$30 d/m/100 cm^2	$\leq$3 d/m/100 cm^2

*The number of samples considered in deriving an average shall include at least 10 samples and there shall be at least one sample from each square meter of the projected surface area.

Areas that could not be made to meet noncontamination zone specifications are called "controlled zones" and, in such areas, when the orange paint shows, the surface will have to be repainted. Most of these were "contamination zones" before the plutonium release and are normally more strictly controlled than required because of the sealed-in radioactivity. For this reason, restrictions associated with the "controlled zone" designation are not troublesome.

During all phases of decontamination frequent smearing and probing were required to evaluate the decontamination methods used and to indicate progress, particularly during the later stages when it was desirable to concentrate only on the "hot" areas. Complete survey of an area requires one smear and four probes, with a gas flow α meter, per square meter of projected surface. Usually, however, the surveys were more thorough than

this. As an example, for one complete survey of the penthouse ceiling, which has a projected surface area of about 420 sq meters, a total of 2084 smears and 7263 probe readings were taken. This averages about four smears and 15 probe readings per square meter, or 4 times the specified amount. A survey of the penthouse ceiling required approximately 16 man-days of effort, and the equivalent of four or five complete surveys was made during the decontamination of the penthouse ceiling alone.

5.4.3 Decontamination Procedures

Strict rules for entering and leaving contaminated areas were set up to prevent the spread of contamination into clean areas. Protective clothing required for contaminated zones was two sets of coveralls, two pairs of shoe covers, two pairs of rubber gloves, an assault mask, and hood, with wrists and ankles sealed with masking tape. When leaving contaminated zones, personnel were directed through check points where successive layers of clothing were removed and personnel were monitored. Personnel then showered and received a final check for contamination before they were released. Frequent monitoring of the change house area indicated that little or no contamination was spread into clean zones.

Before decontamination work was begun on each area, easily removable equipment was wrapped in plastic and moved to an outside decontamination pad. Equipment and supplies of low value were removed to the "burial ground" for disposal.

The first step in decontamination was the removal of gross amounts of contamination by vacuuming or sponging to prevent spread of activity. When a large amount of dust was present, vacuuming proved to be valuable, but it was still necessary to sponge the area to remove additional amounts of easily transferable contamination. Sponging alone proved to be just as effective as vacuuming and sponging. During the sponging operation the amount of water used was limited, and sponges were replaced frequently. Sponging of instrument tubing, wireways, instrument racks, and electrical panels, as well as of flat surfaces, proved effective.

The second step in the decontamination procedure was to thoroughly scrub all surfaces with detergent, water, and stiff fiber brushes followed by thorough water rinses. Several treatments of this type were usually required to remove the remainder of the transferable contamination.

The most difficult part of the decontamination procedure was the removal of nontransferable contamination, >800,000 d/m/100 sq cm in some places, that had become fixed to concrete, metal, and painted surfaces. The use of chemical reagents, such as dilute hydrochloric acid on concrete and dilute nitric acid on stainless steel, removed this fixed activity in some cases. However, it was generally necessary to mechanically remove a layer of the contaminated surface before it would meet the contamination tolerance limits. Relatively smooth concrete floors such as existed in the control room were ground with terrazzo floor grinding machines to remove a layer of concrete approximately 1/16 in. thick and the remaining "hot" spots were chipped out. In areas where the concrete floors were pitted, as in the makeup area, it was necessary to chip out almost the entire floor and pour a new one. Painted surfaces were decontaminated by removing successive layers of paint, in many cases down to bare metal, until acceptable contamination levels were reached. Unpainted stainless steel surfaces were partially decontaminated with dilute nitric acid and then scrubbed with steel wool. Unpainted metal surfaces other than stainless steel were particularly hard to decontaminate, and had to be sanded and ground almost to a polish before specifications were met. In many cases it was less expensive to cut out sections of contaminated piping and replace with new material. The decontamination treatments applied to various types of surfaces are summarized in Table 5.1.

5.4.4 Decontamination of Specific Areas

Decontamination of the penthouse, cold solution makeup area, and pipe tunnel are discussed below to illustrate the variety of problems encountered during building cleanup.

a. Penthouse. The most heavily contaminated building area outside the processing cells was the penthouse, an enclosure 165 ft long, 30 ft wide, and 30 ft from floor to ceiling, constructed of Q-type insulated metal panels and located on top of the cells. Here are located a bridge crane for moving equipment into and out of the cells, instrument transmitters mounted on special racks, miscellaneous pieces of process equipment which are either nonradioactive or are in shielded cubicles, and a large amount of instrument tubing, process piping, and piping for water, steam, and air services. Figure 5.1 is an over-all view of the penthouse showing the complex surfaces that had to be

Table 5.1. Summary of Decontamination Treatments for Various Surfaces

Surface	Character of Contamination	Primary Decontamination Treatment	Cleanup Rate, sq ft/ man-hr	Other Decontamination Treatment
All (walls, floor, ceiling)	Transferable	Scrubbed with detergent and water and brush or sponge	27	Dusty areas vacuumed
Painted metal (walls and ceiling)	Fixed	Paint removed with paint remover and scrapers, and surface scrubbed with soap and water	4	Outer layer of paint removed with sandpaper
Concrete (floor)	Fixed	Ground with terrazzo floor grinding machine	5	"Hot" spots chipped out and vertical surfaces washed with dilute hydrochloric acid
Bare metal (S. S. piping and tanks)	Fixed	Rinsed with dilute nitric acid and scrubbed with steel wool	-	Surfaces abraded with emery paper
Bare metal (other than S. S.)	Fixed	Abraded with emery paper or ground to remove pits	-	-
Lead shielding	Fixed	Rinsed with dilute nitric acid	-	-
Oily metal (pumps)	Fixed	Washed with "Gunk," a commercial solvent	-	-

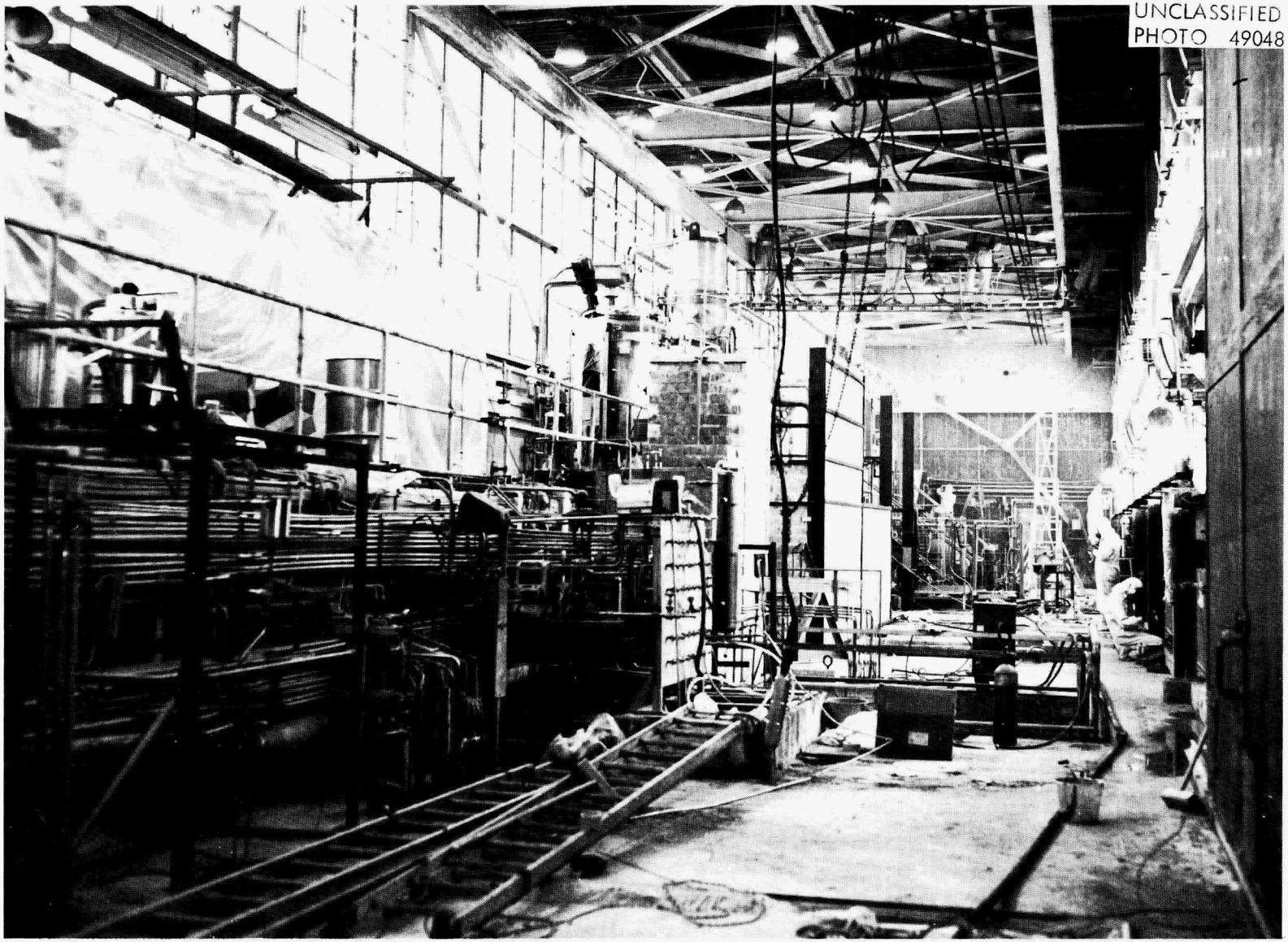

Fig. 5.1. Radiochemical Processing Pilot Plant penthouse during decontamination after explosion.

decontaminated. The penthouse, especially the ceiling (Fig. 5.2) was the most difficult part of the building to decontaminate, and about half of the total building cleanup effort was expended there. Alpha activity had become fixed in the paint, probably during previous programs. After a number of decontamination methods were tried (scrubbing with detergent and water, scraping off loose paint, applying a strippable plastic coating and then stripping it off, and sanding either by hand or with mechanical sanders), the method selected was to remove almost all the paint with paint remover and scrapers and then scrub with detergent and water. Some I beams on the crane supports were removed to a remote decontamination area where they were decontaminated by grinding to a polish. However, this method could not be used in the penthouse because of the danger to personnel of handling portable grinding machines while they were wearing bulky protective clothing, slippery plastic shoe covers, and vision-limiting masks and because of the tendency of grinding operations to spread the contamination by dispersing it in the air.

Three or four men per shift worked three 8-hr shifts per day for about two weeks to decontaminate the instrument transmitters (Fig. 5.3). Because of the materials of construction, copper and mild steel, no acid could be used and the transmitters were decontaminated by scrubbing with detergent and water. The entire racks were wrapped in plastic sheets (upper left part of Fig. 5.1) to decrease the possibility of recontamination resulting from cleanup in other penthouse areas.

The concrete floor of the penthouse was unsuccessfully scrubbed with detergent and water and with dilute acid. It was necessary to remove about 1/8 in. of concrete with a rotary scarifier and then chip out "hot spots" before the contamination was decreased to the point that sealing in the remaining activity with a 2-in. layer of concrete was acceptable.

b. Cold Solution Makeup Area. This area (Fig. 5.4) presented two special problems, a concrete floor badly pitted from acid spills and heavy traffic, and mild steel pipes for plant services (air, water, and steam). After the pumps were removed, the tanks were suspended from the ceiling, the tank support legs were cut off, and the concrete floor was chipped out with jack-hammers. A new floor was poured and a stainless steel floor liner was installed before the tanks and pumps were again in-

Fig. 5.2. Radiochemical Processing Pilot Plant penthouse ceiling during decontamination after explosion.

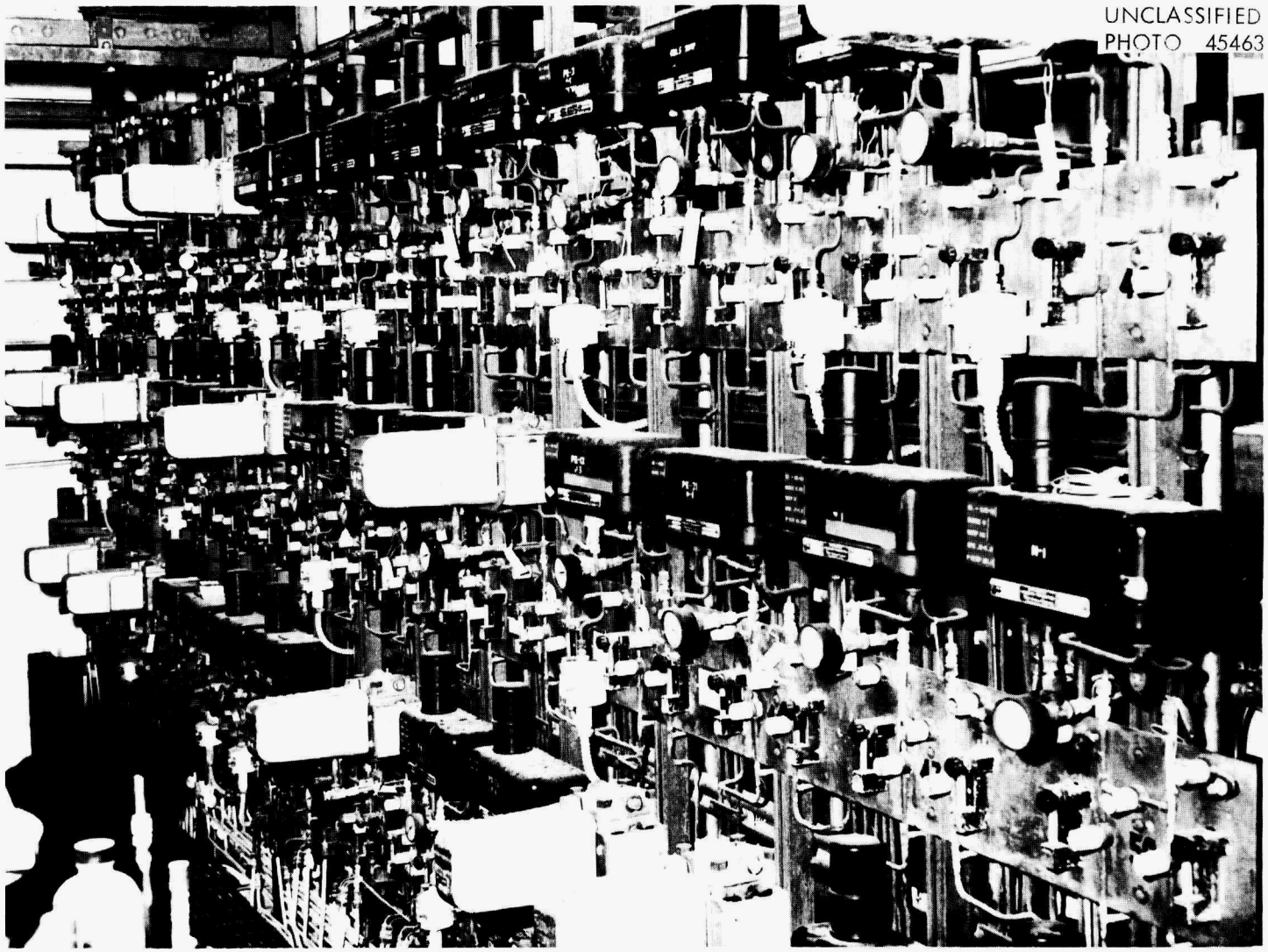

Fig. 5.3. Power reactor fuels processing pilot plant first cycle instrument transmitter rack.

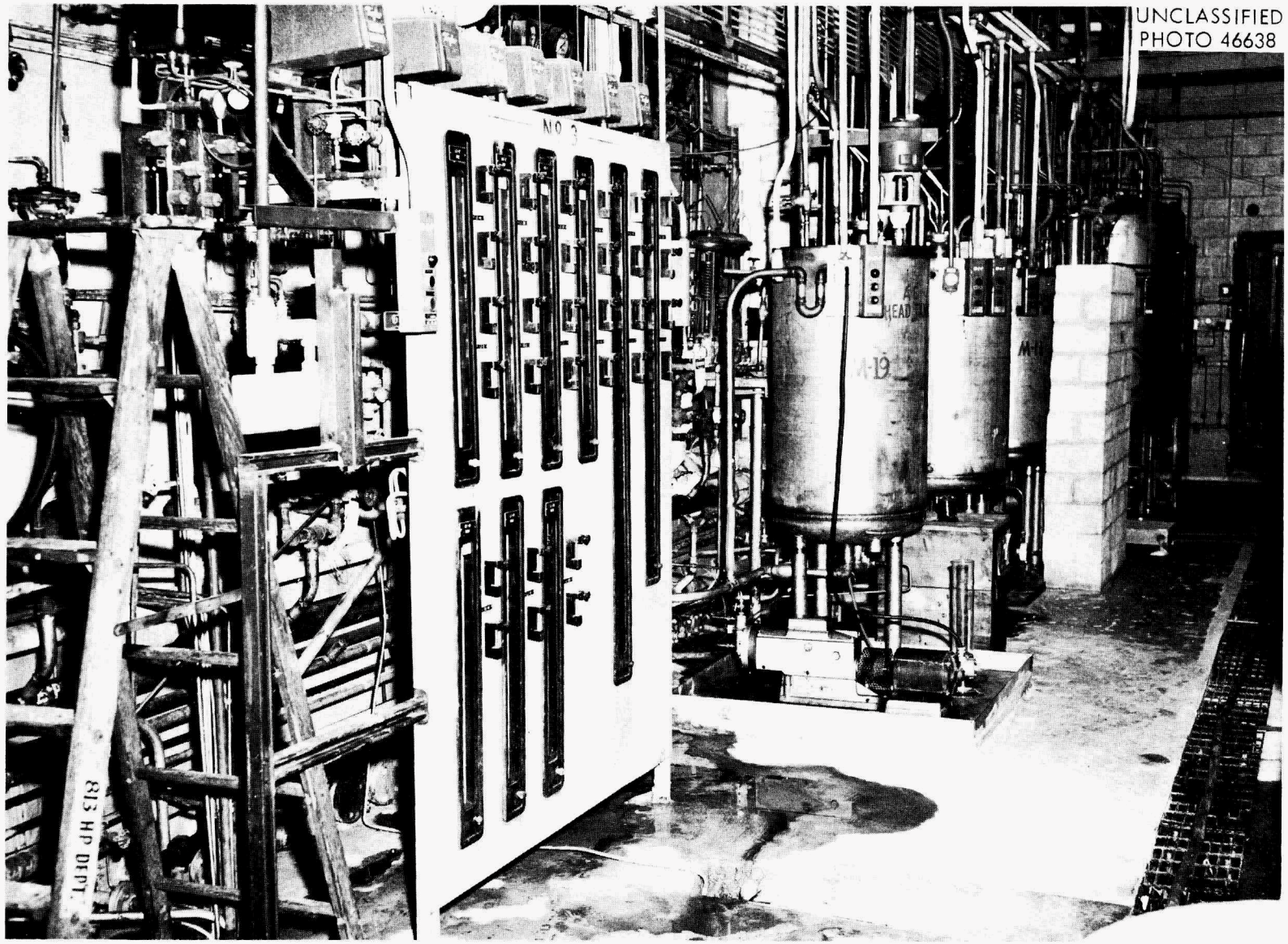

Fig. 5.4. Power reactor fuels processing pilot plant cold solution makeup area.

stalled. Figure 5.5 shows this area during installation of the new floor.

Many of the mild steel pipes could be decontaminated only by grinding to a polish and it was usually cheaper to remove these pipes and install new ones.

c. Pipe Tunnel. The pipe tunnel (Fig. 5.6) is located below the makeup area and adjacent to the processing cells. The surface of the pumps and pulser drive units were coated with contaminated grease and oil, which was removed with a commercial solvent called "Gunk."

5.4.5 Equipment Flushing

Because the material balance for the 27 kg of plutonium processed during the plutonium 240 programs was minus 4,570 ± 3,250 g, the solutions used to flush process equipment were poisoned with either boric acid or sodium borate, and cylindrical plugs were inserted in sumps in cells 6 and 7 as a precaution against criticality.

The ruptured evaporator was flushed in April, 1960. It was expected that little plutonium would be found in this equipment since it had been treated with large amounts of a variety of solutions during the decontamination program. Two 500-liter flushes with 0.1 M boric acid solution yielded 9 g of plutonium. A flush with 500 liters of 6 M nitric acid solution containing 0.1 M boric acid and 0.01 M fluoride yielded about 100 g of plutonium. Flushing of cell equipment was discontinued pending improvement of building containment capability.

Installation of suitable containment and improvements to the vessel off-gas system were completed in August, 1960, and flushing of process equipment was resumed. A solution of 2 M nitric acid, 0.02 M fluoride, and 0.04 M boron was used for all flushing after August.

Of about 1100 g of plutonium flushed from the process equipment, only 10 g was found in equipment that is not a part of the ruptured evaporator. About 700 g was flushed from the steam stripper, P-5, and about 400 g was flushed from the product catch tank, P-3. Catch tank P-3 had held the Turco 4501 solution drained from the evaporator just before the explosion. This solution had been sampled and found to contain about 20 g of plutonium. The Turco Decon 4501 solution was transferred to waste, and the tank was flushed with water (containing sodium borate), which was also

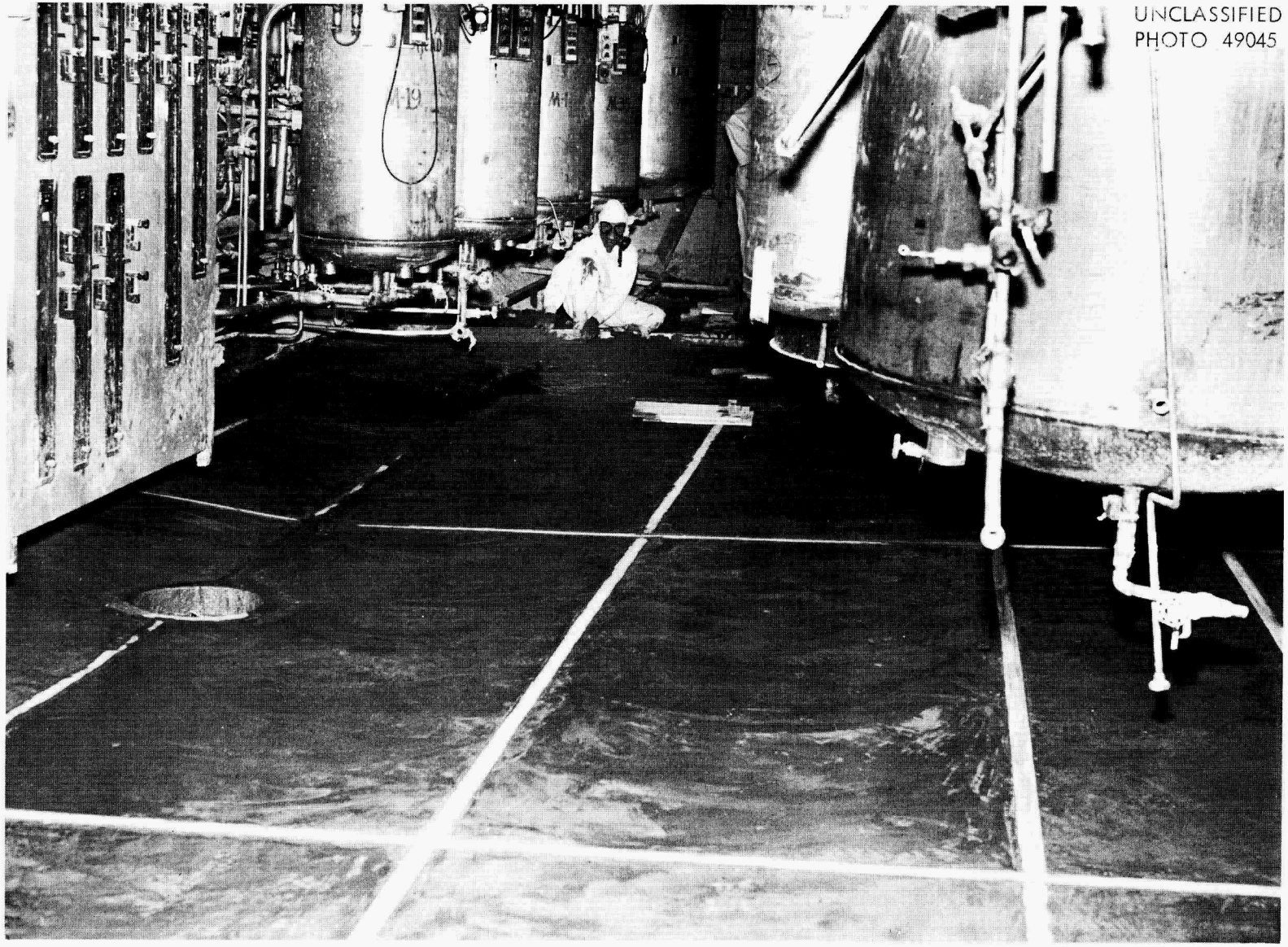

Fig. 5.5. Power reactor fuels processing pilot plant cold solution makeup area during decontamination after explosion.

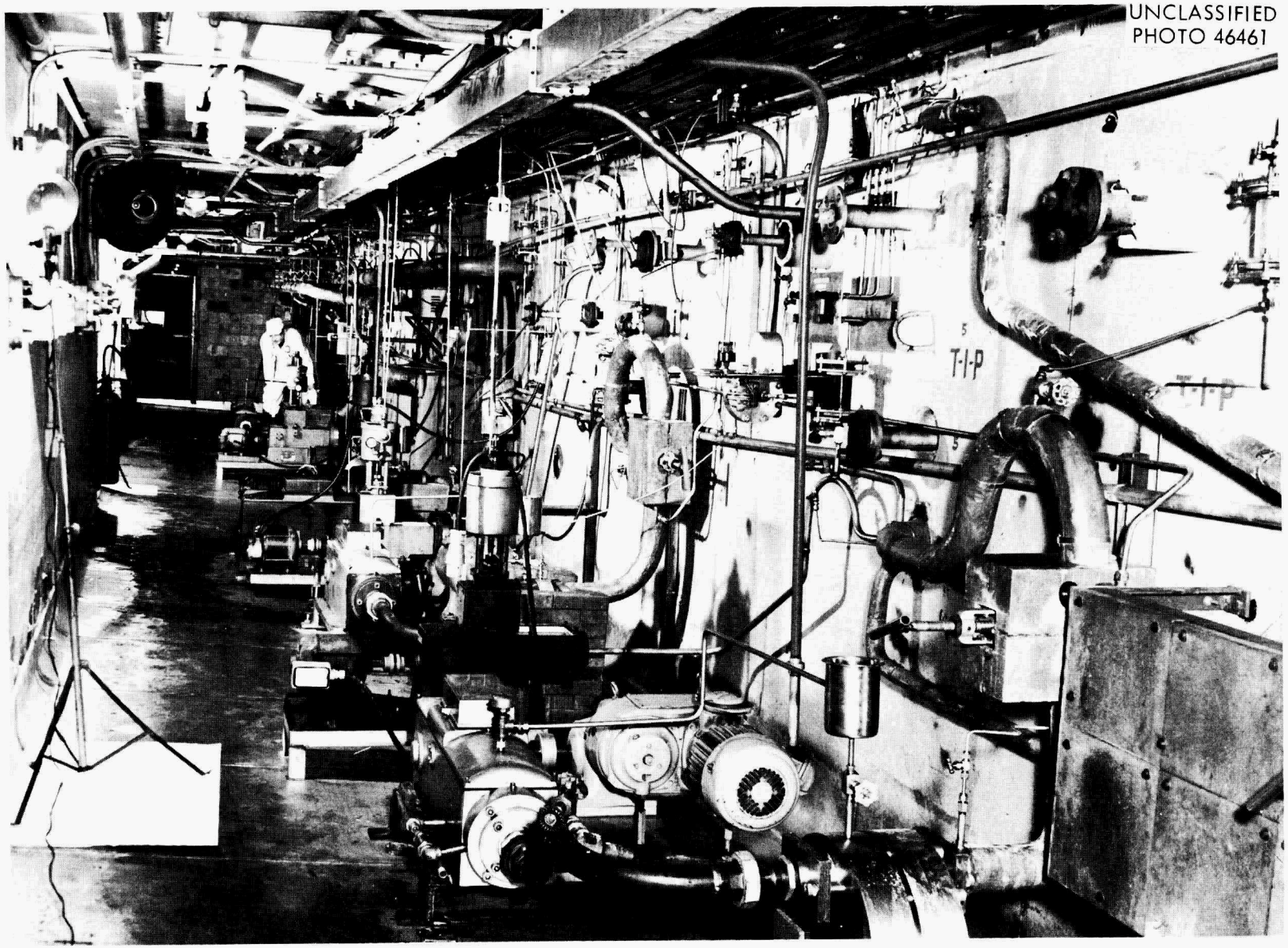

Fig. 5.6. Power reactor fuels processing pilot plant pipe tunnel.

transferred to waste. Subsequent flushing with 2 M nitric acid, 0.02 M fluoride, and 0.04 M boron resulted in the recovery of the 400 g of plutonium.

Two strip column-evaporator combinations were operated in parallel during most of 1959. After the plutonium-240 programs, during a short processing campaign which used only the evaporator not involved in the explosion, the plutonium material balance showed a gain of 170 g. This other evaporator was decontaminated and repaired prior to the explosion but no plutonium was found. Only 3 g was flushed from this evaporator during the August equipment flush.

At present, there is no explanation of why plutonium collected preferentially in steam stripper P-5 during the plutonium-240 processing campaign.

5.4.6 Decontamination of Surfaces in Cells 6 and 7

Cleanup of cells 6 and 7 was started after internal flushing of process equipment was completed. An enclosure made of plywood and transparent plastic was fitted into the cell 6 doorway (which now opens into a secondary containment area) on the metal grating over tanks P-3 and P-4. Most of the cell area can be observed from this enclosure. Thus guessing about the condition of the cell has been eliminated and planning of cleanup operations has been simplified. The cell surfaces in the vicinity of the ruptured evaporator were washed by use of a hose which was directed at the surfaces by an operator who held the hose nozzle through gloves connected to the observation enclosure.

When the observation enclosure was first installed the intensity of penetrating radiation at the front of the enclosure (farthest into the cell) was 400 mr/hr. The hosing operation caused this radiation level to decrease to 200 mr/hr.

Personnel entered the cell and cleaned the debris from in front of the evaporator subcell. Concrete blocks, lead bricks, and sand (from the chipped concrete blocks) were put into 55-gallon drums which were removed from the cell and buried.

The level of penetrating radiation in the observation enclosure decreased to 80 mr/hr. More washing from the observation enclosure reduced the background to 60 mr/hr.

Personnel again entered the cell and washed down the surfaces which could not be washed from the observation enclosure.

The level of penetrating radiation in the enclosure on Nov. 10, 1960, was 60 mr/hr and the levels of penetrating radiation in the cell had been reduced to the point that working time was limited by the physical endurance of personnel working in impermeable plastic suits. The general background in the cells was about 50 mr/hr and the maximum level found with a hard shelled cutie-pie was 2 r/hr.

5.4.7 Results of Decontamination

Most of Bldg. 3019 is probably less contaminated now than it has been at any time in the last 10 to 15 years. Recent surveys made with a portable gas-purged proportional counter were the most sensitive and thorough surveys ever made in the building. Contamination that could not be removed by reasonable means has been bonded in place and covered with either paint or concrete in such a way that a bright color will show before any of the bonded-in activity becomes uncovered. Decontamination of Room 211 and processing cells 6 and 7 was not completed at the time of this report.

About 3500 man-days of effort have been spent on decontamination, not including surveys or maintenance. The only area decontaminated sufficiently to be classified as a noncontamination zone without application of a fixing agent was the offices. Table 5.2 shows the results of the decontamination program. The protective clothing requirements listed are for normal operations only. Drilling holes into surfaces which contain sealed-in activity will require additional protective clothing, and a variety of other unusual operations will require various amounts of personnel protective equipment.

6.0 PERSONNEL MONITORING

No one received more than 2% of the maximum permissible body burden[5] of any radionuclide or more than the permissible dose from external sources of ionizing radiation[6] at the time of the incident or during subsequent cleanup operations. Analyses of more than 300 urine and fecal specimens and six measurements in the in vivo counter (IVRM) were made to monitor internal exposure. The ORNL badge-meters, which are worn at all times by

Table 5.2. Results of Decontamination Program in Completed Areas

Area	Man-days Expended	Final α Contamination, d/m/100 cm²				Fixing Agent		Protective Clothing Required[a]
		Transferable		Nontransferable				
		Avg	Max	Avg	Max	On Floor	On Walls and Ceiling	
Attic	59	7	330	~300	3000	3 coats of paint	3 coats of paint	Minimum
Offices	73	3	18	-	300	None	None	None
Room 100	202	8	128	~300	3000	Tile	3 coats of paint	None
Pipe tunnel	180	18	228	~300	3000	None	4 coats of paint	Maximum
Sampling gallery	181	15	304	~300	3000	2 in. of concrete	4 coats of paint	Lab coat and shoe covers
Makeup	212	25	254	~300	3000	6 in. of concrete	4 coats of paint	Lab coat and shoe covers
Penthouse	1360							Maximum
a. Wall and floor		20	250	300	3000	2 in. of concrete	4 coats of paint	
b. Ceiling		45	500	1100	3000		4 coats of paint	
Cell 3[b]	400	40	264	10^4	$>10^5$	None	4 coats of paint	Maximum
Basement	100	20	300	~300	1000	4 coats of paint	4 coats of paint	Maximum

[a]Minimum = 1 pair of coveralls, shoe covers, and gloves; maximum = assault mask and 2 pairs each of coveralls, shoe covers, and gloves.

[b]Radioactivity is all uranium-233 and daughters.

all employees and which are routinely analyzed every 3 months, were used to monitor exposure of personnel to penetrating radiation. In addition, special film badges and electroscope dosimeters were worn by all personnel who worked in areas suspected of containing measurable amounts of penetrating radiation.

6.1 Internal Exposure

The Applied Health Physics Bio-Assay Group analyzed 327 urine specimens from 242 people and 83 fecal samples from 58 people between November 20 and December 31, 1959. This was not many more analyses than usual for the Bio-Assay Group, but nearly all were concerned with the pilot plant incident. In vivo measurements were made of six people, three of whom, because of their movements on the night of the incident, were potentially exposed to high plutonium contamination and the other three, because of long histories of working around plutonium, to be sure that it would be permissible for them to be actively involved in cleanup and decontamination.

People associated with the incident in any way submitted urine specimens immediately after the incident, and those who entered the highly contaminated areas the night of the incident were asked to submit fecal samples. Everyone who was actively engaged in cleanup operations submitted weekly urine specimens, and anyone who became more than slightly contaminated submitted a urine specimen, which was collected in the 24-hr period following his being contaminated. If any of the test results gave cause for concern, the person involved was removed from all contact with the contamination until it was ascertained that his resuming cleanup operations presented no special hazard to him.

6.2 Exposure from External Sources

Exposure of personnel to external sources of ionizing radiation is monitored by the ORNL badge-meter,[7] which is worn at all times by all Laboratory personnel. Normally, this badge-meter is analyzed every 3 months. When a person works in areas known to contain sources of ionizing radiation, he also wears pocket dosimeters that are analyzed daily and a special film badge-meter that is analyzed each week. The special film badge meter is used to aid in the administrative control of personnel exposures. Pocket dosimeters are used only as indicators of

accumulated doses. If the meters go off-scale (over 200 mrad) or if the total of daily readings exceeds half the permissible quarterly dose, the ORNL badge-meter is analyzed immediately.

6.3 Precautions against Overexposure

The radiation control program during cleanup and decontamination was not limited to monitoring the amount of exposure of personnel. Such exposure was predicted and limited during the entire period. Personnel monitoring techniques were used to check the effectiveness of the control program.

Protective measures to control personnel exposure to α contamination were described in Sect. 5.4.3. Areas were continually surveyed for sources of ionizing radiation. Before anyone entered an area he was made aware of the intensity and location of any fields of ionizing radiation in the area and was told how long he would be allowed to remain in the area. Personnel working in areas that contained sources of ionizing radiation were required to periodically read pocket dosimeters, which they wore in addition to the pencil dosimeters, special film badges, and ORNL badge-meters.

6.4 Results

The effectiveness of the control measures used is illustrated by the exposure record since the explosion. The data available show that no one received an overexposure, either internal or external. During the decontamination program (through June, 1960) the highest 3-month external exposure for any individual was 705 mrem. Three other people received dosages in excess of 600 mrem in a quarter, and four received dosages between 500 and 600 mrem; all others were less than 500 mrem per quarter. The 6-month external exposures (including the quarters above) were: one man, 1070 mrem; three men, 900-1000 mrem each; three men from 800 to 900 mrem; and nine men from 700 to 800 mrem; all the rest received less than 700 mrem total in 6 months.

There were no internal overexposures. An operator who was exposed the night of the explosion and a lead burner who was exposed during decontamination received the highest internal exposures: about 2% of the maximum allowable body burden of plutonium.

7.0 CONTAINMENT OF RADIOACTIVE OPERATIONS

The general philosophy and standards adopted for containment or confinement of reactors have been applied to all radioactive operations at Oak Ridge National Laboratory. Radiochemical pilot plants, "hot" laboratories, and experimental setups, as well as ORNL reactors, must be designed and operated so as to preclude discharge into the Laboratory area and its environment of concentrations or amounts of radioactive materials that would be injurious to health or would interfere with other Laboratory programs. The maximum credible accident must be contained or confined, and two lines of defense must be present to prevent escape of radioactive materials via waste streams. A formal hazards report must be submitted to an appropriate Laboratory review committee[2] for operations involving more than 1 g of plutonium (or equivalent of other α emitters) or more than 1000 curies of β-γ emitters.

7.1 Chemical Technology Division Containment Program

All Chemical Technology Division facilities and operations have been reviewed to determine the maximum credible hazards and the extent of modifications that would be required to provide a degree of protection consistent with the potential hazards. In general, massive cell shielding can provide primary containment that is capable of maintaining its integrity through any credible incident, and the circumscribing building structure can serve as the secondary line of containment capable of closure for evacuation, on emergency signal, to a pressure below atmospheric. The provision of adequate scrubbing and filter systems on off-gas lines and the necessary sampled hold tanks or monitored diversion boxes in liquid waste disposal systems must be such that inadvertent escape to the environment through these penetrations would require a minimum of two coincidental and unrelated failures.

7.1.1 Primary Containment

The massive shielding required for attenuation of penetrating radiations in most Chemical Technology Division radiochemical operations is suitable for a primary containment shell. The physical strength of the concrete, iron, or lead shielding may be relied on to withstand the pressure of chemical decompositions. Violently energetic air-organic explosions can be prevented by use of alarm organic vapor detection instrumentation and manually operated water spray systems.

The degree of leak tightness of the primary containment shell depends on the time-pressure integral after an incident. This integral, in turn, is a function of the rate of explosion suppression and normal ventilation exhaust capacity for the cell. Computations based on pessimistic assumptions have suggested the following criteria for the primary containment shell:

1. Negative pressure in cell: 1 in. water
2. Ventilation exhaust capacity: 0.1 cell volume/min
3. Capability of withstanding internal shock pressure of 900 lb/sq ft
4. Ventilation inlet to be closed on emergency signal and ventilation exhaust provided with absolute filters
5. Inleakage: 10^{-2} cell volume/min at 2 in. water differential pressure
6. Penetrated faces of primary enclosure surrounded by a secondary containment shell.

7.1.2 Secondary Containment

The secondary containment is intended to ensure that any air-borne radioactivity that might escape from the primary containment will not be dispersed to the environment. It is probable that some leaks will exist or develop in the building, since it is economically and operationally impossible to provide an absolutely leak-tight structure. Air-borne radioactive materials could flow into the lee vacuum induced by a strong wind. This eventuality could be prevented by providing for reduction of building air pressure to approximately 0.3 in. water below atmospheric promptly on emergency signal.

Necessary openings in the secondary enclosure such as access doors must be through air locks.

7.1.3 Off-Gas Disposal

Under the revised containment policy, five types of off-gas, other than building ventilation, are recognized. These are, in order of increasing potential activity content, secondary containment area ventilation exhaust, laboratory hood off-gases, cell or primary containment area off-gas (COG), vessel off-gas (VOG), and dissolver off-gas (DOG). During emergencies the exhaust ventilation air from secondary containment areas

must be passed through single roughing and absolute filters or directly into primary containment areas. The other four types of off-gases should be treated on a continuous basis, i. e., during normal as well as emergency operation.

Hood off-gases and primary containment (cell) off-gas will be filtered through roughing and absolute filters and then discharged to the environment through a plant stack. Cell off-gas from the Volatility Pilot Plant will, in addition, be provided with a caustic scrubber to protect the filters in the event of a fluorine or hydrogen fluoride leak from the process equipment.

Dissolver and vessel off-gases will be caustic-scrubbed to remove iodine and corrosive gases such as nitrogen dioxide and then filtered through roughing and absolute filters at the off-gas source. They will then be routed to the 3039 stack, where they will be rescrubbed and refiltered before discharge. A silver-coated copper mesh iodine absorption unit will also be provided at the 3039 stack for further iodine removal when required. Dissolver off-gases will be collected separately and treated further at the source when appropriate; equipment will be provided for safe burning of hydrogen and decomposition of ammonia gas produced in Sulfex and Zirflex decladding operations, respectively, and for removal of excessive quantities of krypton and xenon.

Cell, vessel, and dissolver off-gas will be monitored at the final discharge point; an alarm set to signal release of off-gas at concentrations in excess of 10 MPC will be provided.

7.2 Building 3019 Containment

At the time of the explosion of the intercycle evaporator, Bldg. 3019 was not capable of containing a minor chemical explosion without release of radioactive contamination from the building. From the time this building was first built in 1943 until the evaporator exploded, controlled air flow from the occupied or low-activity areas through unoccupied high-activity areas to an off-gas treatment facility and discharge stack was the only means used to limit the spread of air-borne activity. There have been instances when air-borne activity levels in the building have been above tolerance for short times, but these instances were of little consequence and changes in operating procedures were sufficient to pre-

vent recurrence. As plants became larger or more complicated and as radioactivity levels increased, the off-gas system was modified, but the use of controlled air flow for air-borne activity control was retained. Figure 7.1 shows the general air flow pattern as it existed just prior to the explosion.

The application of the new containment criteria to Bldg. 3019 has shown the need for structural changes to the building, modifications to the off-gas and ventilation systems (including improved instrumentation), and additional equipment for fire and explosion protection. Proposed modifications are described below.

7.2.1 Proposed Structural Modifications

Prior to cell cleanup operations a concrete block wall was installed under the sampling gallery parallel to the cells. This wall will become a part of the building secondary containment structure. The Bldg. 3019 cell structure and building superstructure will be renovated to guarantee the degree of leak tightness stipulated under the new containment criteria. Metal siding will be installed over the existing window areas of the penthouse, sampling gallery, and east dock area of the building. The exterior surfaces of all primary containment areas (cells and basement) and of all secondary containment areas (operating gallery, attic, penthouse, sampling gallery, and east dock area) are to be sealed to minimize air leakage. Every effort will be made to provide a negative pressure in excess of 1 in. water in all cells during operation and to ensure the rapid attainment of a negative pressure in excess of 0.3 in. water in all secondary containment areas during an emergency. The pipe tunnel and east dock area (plutonium handling) will be kept at 0.3 in. water negative pressure at all times. Double-door air-lock systems will be installed on all entrances to primary and secondary containment areas, and the cell doors will be designed to withstand an explosive shock pressure of 900 psf. A new secondary containment partition parallel to the cells will be installed through the full length of the attic space above the operating area.

7.2.2 Proposed Off-gas and Ventilation Modifications

Three major additions have been proposed for the Bldg. 3019 off-gas systems. These include (1) installation of a bank of filters and associated fans and ducting for filtering the hot analytical facility cell off-gas

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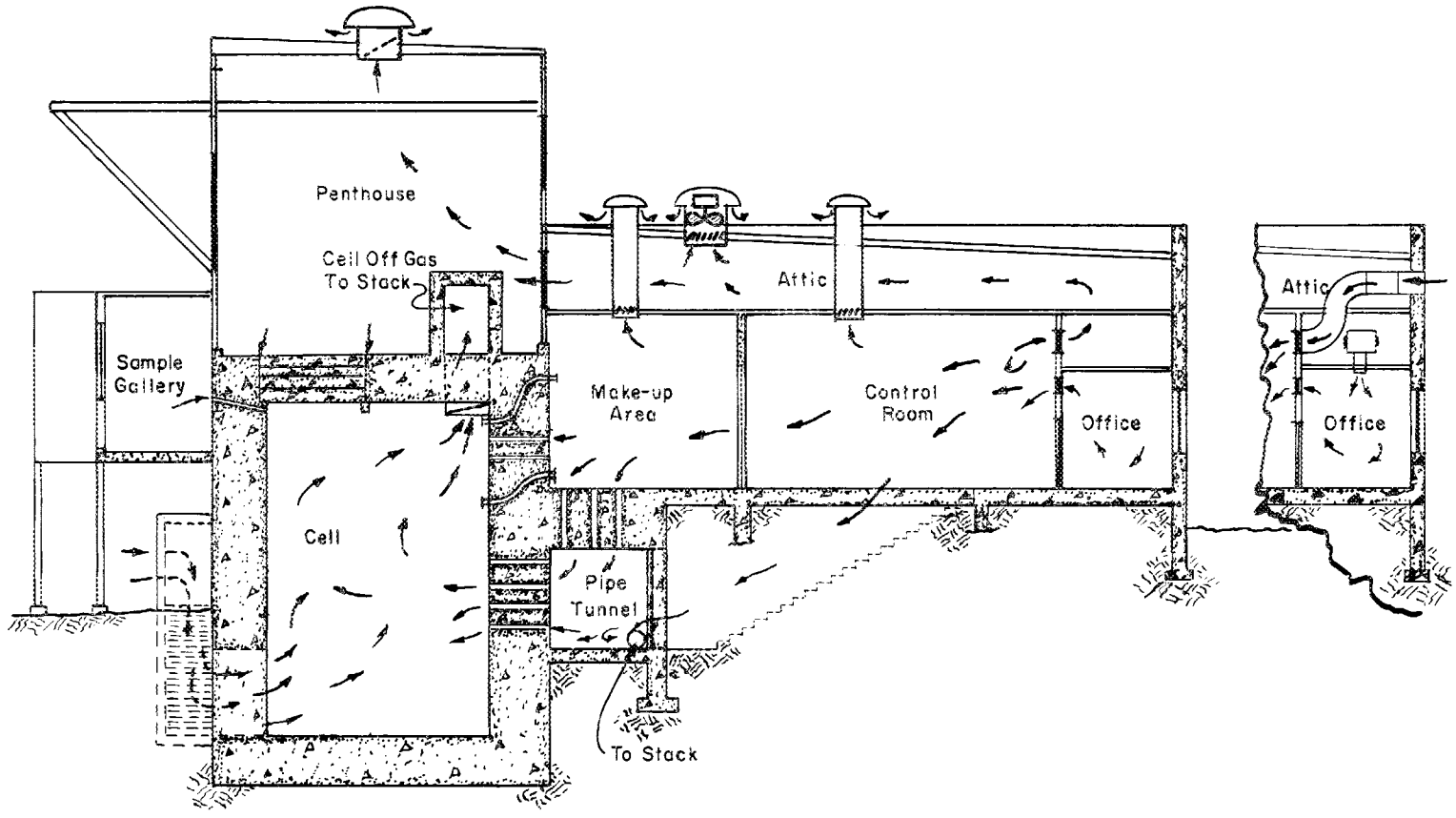

Fig. 7.1. Radiochemical Processing Pilot Plant air flow prior to the evaporator explosion.

prior to discharge to the 3020 stack; (2) installation of a caustic scrubber and filters for the dissolver and vessel off-gas prior to its release to the 10-in. stainless steel line to the 3039 stack; and (3) installation of a potassium hydroxide scrubber for the Volatility Pilot Plant off-gas from cells 1 and 2.

Changes in the existing primary containment cell off-gas system at Bldg. 3019 include (1) revision of cell air intake systems, (2) alteration of cell exhaust system to the cell off-gas header, and (3) upgrading of the instrumentation for the cell off-gas handling equipment in the 3020 stack area. Similar air control systems for the basement, pipe tunnel, and the east dock plutonium-handling area will be provided. All primary containment air will be supplied from the Bldg. 3019 penthouse.

Except during periods of process equipment maintenance or construction, when air will enter the cell through the 9-by 9-ft roof plug openings, all cell air inlets will pass from the penthouse to the cell through new ducts, each of which contains a filter, a back-flow preventor, and a flow control valve. Each cell exhaust will be equipped with a closure device, a flow meter, perhaps a back-flow preventor, and a flame arrestor.

Revised instrumentation at the 3020 stack area will guarantee automatic switching of the cell exhaust load from the electrically driven fan to the steam-driven emergency unit in case of a power failure or low vacuum in the processing or high-level analytical cells. It will also ensure a constant vacuum in the cell off-gas header at the cells by providing fan suction control valves and associated instrumentation and provide for measurement of the total air flow and filter pressure drop with appropriate reporting of these data within Bldg. 3019.

Ventilation changes for the remainder of Bldg. 3019 will be minor. The sampling gallery will be ventilated by air recirculated from the penthouse. Closure devices will be installed on all normally open air intakes in the secondary area. Exhaust ports for the offices and control rooms (which are outside the secondary containment area) will be installed in the roof vents leaving the ceiling of the control room area so that normal ventilation can be maintained within these areas during an emergency shutdown.

In all secondary containment areas air will be evacuated to a negative pressure of 0.3 in. water during an emergency by closing the air intakes on the secondary containment area, thereby allowing the primary ventilation system to evacuate the secondary area. When the secondary containment vacuum reaches 0.3 in. water, pressure switches on the air passages that connect the penthouse and the cells will be closed and prevent excessive vacuum in the penthouse. The vacuum established in the penthouse area will apply to all other secondary containment areas, since all will communicate with the penthouse via open ventilation louvers.

7.2.3 Proposed Instrumentation Additions

The proposed ventilation revisions described above will be controlled from a new central emergency control panelboard to be located in the Bldg. 3019 control room area. Pressures within all primary containment areas, the pipe tunnel, and the penthouse will be recorded continuously on this panelboard. The total flow to the cell exhaust fans and the pressure differential across the filters will also be recorded. Indicating instruments will be provided for the air exhaust flow from each primary containment area and for the pressure within all other secondary containment areas. All closure devices on air intake systems to secondary containment areas, and all roof exhaust ports from the control room area will be remotely operated from this panelboard. The output of all process waste, cell off-gas, and vessel off-gas monitors and of all secondary containment area monitrons and constant air monitors (CAM's) will be recorded. All such recording and indicating instruments will be equipped with appropriate alarms.

An important feature of the proposed emergency control panel is the emergency "scram" system which will automatically actuate the appropriate closures required to effect emergency ventilation conditions. Automatic change-over is planned in the event of an explosion in a cell and manual operation will be possible.

7.2.4 Fire and Explosion Protection

All primary containment areas which handle solvents or other inflammables (cells 3, 5, 6, and 7 and the basement-pipe tunnel area) will be equipped with a manually operated fire protection water spray system, "rate-of-temperature-rise" fire detection instrumentation, and infrared

organic vapor detection instrumentation. The spraydown system for each 19 by 20 by 27 ft cell will consist of 16 nozzles, each with a capacity of 10-12 gpm. The fire detection instrumentation would sound an alarm on the emergency control panel and will automatically shut down the air intake to the affected cell. The output from the organic vapor detection instrumentation will be recorded at the emergency panel and will be instrumented to sound an alarm.

8.0 CONCLUSIONS

The release of plutonium from Bldg. 3019 and resulting contamination of the cell 6, Bldg. 3019, and adjacent facilities was the only significant result of an explosion of chemical origin which occurred in the Radiochemical Processing Pilot Plant on Nov. 20, 1959. Nitrated organic compounds, specifically picric acid, were formed while decontamination operations were in progress using a proprietary decontaminating agent containing phenol, organic acids, and amines. The immediate steps which led to the explosion involved an operational error which resulted in the organics being boiled with concentrated nitric acid. No significant radiation exposure of personnel and limited contamination of property occurred because of the effectiveness of built-in protective systems, especially exhaust ventilation filters; prompt action on the part of operating staff and Health Physics personnel; and the fortunate circumstance that the incident occurred during a period of low-velocity wind on Friday evening, before a week end.

8.1 Plutonium Release

The plutonium α activity that contaminated streets and buildings in the vicinity of the Radiochemical Processing Pilot Plant was released from that building through the cell 6 door. The area originally contaminated was limited because of a light wind and because the fallout rate was rapid. The number of people involved was small because the explosion occurred late at night when the ORNL population was at a minimum. The contamination was confined to a small area because of the prompt and correct actions of plant operators, shift supervisors, and health physicists. The intercycle evaporator had been treated and flushed but still contained plutonium as a nitric acid insoluble compound, probably oxide--about 700 g has been flushed from it since the explosion. Cell 6 door, which was about 20 ft from the evaporator, was blown open as a result of the explosion and provided a direct path for the contamination to escape from the cell.

If cell 6 door had not opened, it is probable that the contamination would have been confined to Bldg. 3019.

Open access holes from processing cells 6 and 7 to the makeup area, pipe tunnel, sampling gallery, and penthouse were the paths by which the plutonium escaped from the cells to contaminate the interior of Bldg. 3019. The pressure rise in the cell after the explosion was probably not severe and probably was of short duration. Simple provisions for plugging the access holes and the sealing of the cell 6 access door (installed in 1946) might have prevented escape of the radioactivity from the cells.

8.2 Steps Leading to the Explosion

The Thorex dissolver was decontaminated in late 1958 by the Turco 4501 process which proved superior to previously used treatments. The Turco process required 4 man-days of effort and $872 for chemicals, produced 700 gal of waste, and decreased the radiation level to 70 mr/hr at contact with the dissolver. The process used for a previous dissolver decontamination required about 100 man-days of effort and $675 for chemicals, produced 3000 gal of waste, and decreased the radiation level to only 2000 mr/hr. On the basis of this test and because of the continued success of the method, the Turco 4501 process or a modification had been used for nearly all major decontamination programs since January, 1959.

Instructions issued by the manufacturer called for treatment at 270°F by Turco Decon 4501 (the boiling point), thorough water rinse, treatment with Turco Decon 4502 at 220°F, thorough water rinse, and passivation by 30% nitric acid at room temperature. The Turco Decon 4502 treatment had no beneficial effect on decontamination and, in fact, caused detrimental deposits of manganese dioxide, which were not easily removed from equipment. The Turco Decon 4502 treatment was dropped from the process by the pilot plant operating group.

The decontamination sequence as normally used in the pilot plant, beginning about March, 1959, consisted of a Turco Decon 4501 treatment at 80-90°C, a water rinse, a 30% nitric acid treatment at 80-90°C, and a final water rinse. The intercycle evaporator was treated with one complete, modified Turco sequence, with hot 25% sodium hydroxide flush and with extensive water rinses, but radiation levels did not decrease. It was decided to make another Turco Decon 4501, water, and acid treatment. Instructions

were written in the program instruction log book to add Turco to the evaporator, heat, drain, and "continue with H^+, H_2O, etc." The shift foreman, following these instructions in which the proper order of the Turco-water-acid treatment was inadvertently rearranged, proceeded with a Turco Decon 4501-acid-water sequence.

This procedure probably would not have been hazardous had not the evaporator retained a "heel" of the Turco decontaminating agent. Because the manually operated evaporator bottom drain valve was located in a high radiation field (it was opened normally only after partial decontamination to flush the evaporator circuit), the remotely operated, side-mounted product valve and line were used for drainage. This valve was elevated sufficiently above the lowest point in the evaporator to allow the retention of 16.5 liters of solution, which in turn contained about 14 liters of "as received" Turco Decon 4501. To this heel, in accordance with the instructions in the program instruction log book, 30% nitric acid was added. Normally this acid is heated just to 80-90°C, but this time it was boiled and the nitric acid was concentrated by drawing off condensate.

The sequence of events which led to the explosion and the release of the activity was:

1. Inadvertent omission of water rinse because of an error in writing program instructions.
2. Incomplete draining of the Turco Decon 4501 from the evaporator.
3. Boiling and concentration of nitric acid which nitrated organic components of the Turco compound to explosively unstable and heat sensitive compounds.
4. Contact of the nitrated organic, in all probability picric acid, with the hot wall of the evaporator heat exchanger.
5. Explosion of nitrated organic, which ruptured the evaporator at its weakest point and dispersed plutonium and zirconium-niobium, probably as insoluble oxides, to the cell interior.
6. Opening of the cell 6 door (at grade) by the weak pressure wave developed by the explosion allowed contaminated cell air to escape. Openings in the cell structure provided passageways for contaminated air to reach other parts of Bldg. 3019 and to a limited extent the Graphite Reactor Building.

8.3 Off-gas Filters

The filters in the cell off-gas system (Sect. 3.5.4) retained very efficiently the radioactivity released to the cell off-gas system. All experimental evidence indicates that no plutonium was released to the environment via the cell or vessel off-gas systems and 250 ft stack.

8.4 Area and Building Decontamination

The area of ORNL contaminated with the plutonium dispersed by this accident was small and confined to facilities immediately adjacent to Bldg. 3019, particularly to the south and east. All facilities were usable on Nov. 23, 1959, two days after the release except for the pilot plant portion of Bldg. 3019 and the Graphite Reactor Building. Operations in the Graphite Reactor Building were resumed on Dec. 22, 1959, after thorough decontamination. Building 3019 areas, except for the cells in which the evaporator explosion occurred, are in use or clean enough for use. Operation without restrictions due to contamination in the Volatility Pilot Plant (cells 1 and 2) has been possible since July, 1960.

Because of the indicated presence of a reasonable quantity of plutonium (now known to be approximately 1300 grams) in the equipment and cells where the explosion occurred, decontamination of these areas was delayed until effective but interim secondary containment was installed, this to preclude the possibility of another plutonium dispersing accident during decontamination in the seventeen year old structure. This decontamination is now essentially complete.

8.5 Administrative Action as a Result of the Explosion

The relatively minor explosion in Bldg. 3019, coupled with other incidents of a similar nature at ORNL and other sites, sharply focused attention on problems of protection against serious radioactive material release from accidents in radiochemical operations. It was recognized that many of the high level operations were carried out at ORNL in facilities designed and built seventeen years ago. In the years since the facilities were built, process equipment and techniques were changed and specific activity levels and total quantities of radioactive materials were increased by orders of magnitude, but structures and ventilation systems basically have been altered only slightly in order that a great variety of important development programs might be expedited at a minimum overall cost.

Immediately following the explosion, all high level radiochemical operations at ORNL were closed down, pending extensive studies necessary to define a simple system of secondary containment. In addition, formal hazard reports were prepared for eight of the radiochemical facilities at ORNL. The attention of all personnel to problems of designing and maintaining safe facilities has been assured by the appointment of a Director of Radiation Safety, divisional radiation safety officers and expert review committees for all installations and operations; these functions supplement the long established health physics, safety, fire protection, and medical services of the Laboratory.

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3. "Total Plutonium, Thenoyltrifluoroacetone Extraction Method," ORNL Master Analytical Manual, Method 9 008621.

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6. "Permissible Dose from External Sources of Ionizing Radiation," U. S. Dept. of Commerce, Natl. Bur. Standards, Handbook 59 (September 24, 1954).

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Appendix A. CHRONOLOGY OF EVENTS CONCERNING THE INCIDENT

(All times are approximate)

November 18, 1959 and previous: PRFP equipment washed with water, acid, and water to remove soluble plutonium, etc.

November 18-19, 1959: First Turco Decon 4501-water-acid decontamination series in evaporator.

November 20, 1959, midnight to 3:00 p. m.: Evaporator system flushed with hot 25% NaOH, followed by 1200 liters of water.

3:30 p. m. to 8:45 p. m.: Added 200 liters of Turco Decon 4501 diluted with 62 liters of water; heated 2 hr; drained (except for 16.5 liters below drain elevation); 225 liters drained to P-15.

8:50 p. m. to 9:30 p. m.: Added 270 liters of 30% nitric acid.

9:30 p. m. to 10:58 p. m.: Boiled evaporator contents, distilling off about 200 liters through condenser to catch tank.

10:58 p. m.: Explosion; 48 liters left in evaporator plus scrubber at time of explosion.

10:58 p. m. to 11:00 p. m.: All three plant operators plus foreman in control room at time of blast; all four looked into makeup room, to see room filling with a fog. Foreman sent one operator for gas masks; he asked other two operators to leave building and picked up cutie-pie; no activity detected.

Two operators from Volatility Plant (located immediately east of PRFP) and four people from Hot Analytical Lab came toward Control Room. Foreman sounded evacuation alarm and all except one operator and foreman immediately left. Someone dispatched to locate janitor seen earlier in building lunchroom but apparently he had already left.

Foreman and operator donned gas masks; made quick check and left.

Health Physics surveyor, walking from ORR to Graphite Reactor Building, heard blast. Encountered Reactor Projects Division employee walking on Hillside Avenue toward ORR. Returned to Health Physics truck parked at ORR Building, turned on instruments, and headed for Bldg. 3019 up Hillside Avenue in truck. When opposite cell 6 noted cell door open and unmortared

block wall formerly on street side of door blown over. He surveyed street with Geiger counter; found 15 mr/hr and took smears of street to be counted later. Proceeded to west gate of Bldg. 3019 where evacuees had congregated.

At time of blast Laboratory shift supervisor was in west end of plant near Medical Building; heard blast and entered car. Proceeded immediately to Bldg. 3019. While on way, received radio call for him to go to Bldg. 3019.

11:00 p. m. to 11:30 p. m.: Plant shift supervisor arrived. Found all personnel accounted for. Issued call for guard and fire captains, all available health physicists (3), and shift electrician. Evacuees checked for β-γ activity by surveyor. Little found.

Four building technical supervisors notified by phone. All reported ~20 min later.

Foreman and operators re-entered building in masks to cut off steam and water and otherwise secure operations. Health Physics surveyor re-entered to make survey and read air monitors. Filter papers removed from air monitors.

Health Physics surveyors dispatched to map extent of activity spread to east. (Very low velocity wind from west.) Health Physics surveyors notified Health Physics Division management. Shift supervisor notified Plant security and safety chiefs, who reported ~20 min later.

11:30 p. m. to 12:00 midnight: Building technical supervisors arrived on scene; guard posted at Third Street and Central Avenue to keep out unnecessary personnel; guard posted at Hillside Avenue to require shoe covers beyond this point. Order issued to guards at plant gates to not allow midnight shift to enter or others to leave plant without being given Health Physics check.

Phone call to member of PRFP data group reconfirmed information known earlier in day that book discrepancy of 1200 g of plutonium existed. Air sample filters sent to analytical lab for count and identification; β-γ count made first and principal γ activity found as Zr-95, Nb-95; α count requested and analyst went to different building to make it. Building 3019 personnel field checked for α, but little or none found.

Building re-entered with masks to collect air sample filter from penthouse. Sample read 210 mr/hr β-γ. Sample sent to analytical lab during next period.

Foreman and a supervisor looked into open door to cell 6. Cutie-pie reading at doorway was 7 r/hr. Light poor but supervisor saw unmortared block wall in front of evaporator blown over and evaporator tipped. Piping in evaporator subcell disarranged. Supervisor contaminated to 20 mr/hr β-γ. Health Physics surveyors dispatched to gate to check all people leaving plant.

As a precautionary measure, Metal Recovery and Fission Products Pilot Plant shut down principally to reduce volume of process waste water.

12:00 midnight: Plant gate opened to allow shifts to enter and depart.

November 21, 1959, 12:15 a. m.: Two truckloads of departing 3000 Area workers checked for α, β, and γ, but none found.

12:15 a. m. to 12:30 a. m.: Analytical lab reported 4000-13000 α c/min on 3/4-in. square samples of filter paper from air sample. Alpha pulse height analysis requested. Analyst said he had no recent experience in this analysis but would try anyhow. One centimeter square of penthouse filter almost blocked α counter in analytical lab. A Health Physics surveyor independently found α on shoes. Alpha contamination found on supervisor who had looked into cell 6 and in street.

Started checking on liquid waste streams from plant.

12:30 a. m. to 1:30 a. m.: Analyst reported α activity spectrum looked like U-233. Asked to recheck results. Analyst called in man familiar with apparatus. Only reasonable source of U-233 would be ~36 kg of U-233 located in "ever safe" storage rack in pipe tunnel of Bldg. 3019. Pipe tunnel is a room just below Makeup room and just outside cells. Possibility of criticality considered.

Personnel re-entered building to collect threshold detectors. Detectors sent for counting; no evidence of neutron burst. By 2:00 a. m. criticality incident ruled out as likely event.

About 12:30-1:00 a. m. air monitor gave alarm to LITR. Health Physics surveyor sent to check; found instrument returned to near normal. Activity probably originated from experiment at LITR.

1:30 a. m. to 2:30 a. m.: Hillside Avenue barricaded at Third and Fifth Streets and Fourth Street at Central Avenue.

12:30 a. m.: Sample of process waste stream showed 25 c/min/ml α at diversion box. Sample from manhole 26 at Metal Recovery Building showed 150 c/min/ml α.

2:00 a. m.: Sample at waste treatment plant showed 25 c/min/ml at inlet and 15 c/min/ml at outlet. Water treatment plant shut down and process water diverted to equalization basins. Completed by 2:45 a. m. To minimize storage problems, further process-water-saving measures started.

2:30 a. m. to 4:00 a. m.: Analytical lab concluded α activity was Pu-239. Alpha activity found in Graphite Reactor Building.

4:00 a. m. to 6:00 a. m.: Graphite Reactor shutdown completed and personnel evacuated at 4:35 a. m. LITR and ORR found uncontaminated (later some tracking occurred to ORR, which was cleaned up). Personal shoes on eight occupants of Graphite Reactor Building replaced. Over-all mapping of spread of activity completed.

6:00 a. m. November 21 to 8:00 a. m. November 23: Decided to fix α activity spread with paint and tar. Painted south side of Bldg. 3019 and north side of Bldg. 3022. Painted all objects and pieces of equipment on south side of Bldg. 3019. Painted grass on north side of Hillside Avenue.

Purchased 5000 gal of asphalt and 17 truckloads of gravel. Applied about 6000 gal of asphalt and gravel to roads and roofs of three buildings (3019, 3022, 3025). Changed air conditioning filters for Bldg. 3025. Greatly decreased process water flow throughout Laboratory area to within waste disposal capacity.

Cleaned Bldgs. 3022, 3025, 3042, 3010, 3012, 3044 for occupancy Monday morning, November 23. Building 3005 still required shoe covers.

Prepared and printed 3000 copies of handout for all employees arriving Monday morning identifying areas affected and giving occupancy rules.

Appendix B. HISTORY OF PROCESSING IN BUILDING 3019

The processing cells were constructed in 1943 for the pilot plant demonstration of the Bismuth Phosphate process for separating plutonium from natural uranium fuel. Since then, 63 kg of plutonium, 3 kg of U-235, and 170 kg of U-233 (including reprocessing) have been processed in these cells (Table A.1).

Table A.1. Summary of Building 3019 Operations

Year	Process	Amount Processed
1944	Bismuth Phosphate	250 g of Pu
1945	Bismuth Phosphate	50 g of Pu
1946	-	-
1947	-	-
1948	Redox	45 g of Pu
1949	Redox and Hexone "25"	588 g of Pu plus 2460 g of U-235
1950		1,031 g of Pu
1951	Purex	390 g of Pu
1952	Purex	2,930 g of Pu
1953		-
1954	Thorex	2,300 g of U-233
1955	Thorex	5,100 g of U-233
1956	Thorex	20,500 g of U-233
1957	Thorex	24,100 g of U-233
1958	Thorex	30,000 g of U-233
1959	PRFP	51,600 g of Pu

Appendix C. POWER REACTOR FUEL PROCESSING

During the fall of 1958 revisions and additions were made to the Thorex Pilot Plant (Bldg. 3019) and to the Metal Recovery Plant (Bldg. 3505), and the facilities were connected by transfer lines to obtain the Power Reactor Fuel Processing (PRFP) complex. This complex consists of a fuel element dissolver and a solvent extraction co-decontamination cycle, complete with solvent recovery cycle in Bldg. 3019, and solvent extraction partitioning and second uranium and plutonium cycles, complete with plutonium ion exchange isolation columns and solvent and acid recovery facilities in Bldg. 3505. A dissolver for relatively low activity fuel elements is also provided in Bldg. 3505.

The flowsheet for the Bldg. 3019 solvent extraction co-decontamination cycle is presented in Fig. A.1. Aqueous feed containing plutonium, uranium, and fission products is pumped to the HA extraction column where the uranium and plutonium are extracted from the aqueous into the organic phase. The uranium-plutonium-bearing organic stream then passes through the HS scrubbing column, where it is further decontaminated from fission products, and is fed to two parallel strip column--evaporator circuits. In the columns (HC-1 and HC-2) the uranium and plutonium are stripped from the organic into the aqueous phase, which is in turn concentrated in the evaporators (P-15 and P-62) and collected in catch tanks (P-3 and P-4) for sampling and transfer to Bldg. 3505 where the uranium and plutonium are separated and processed further.

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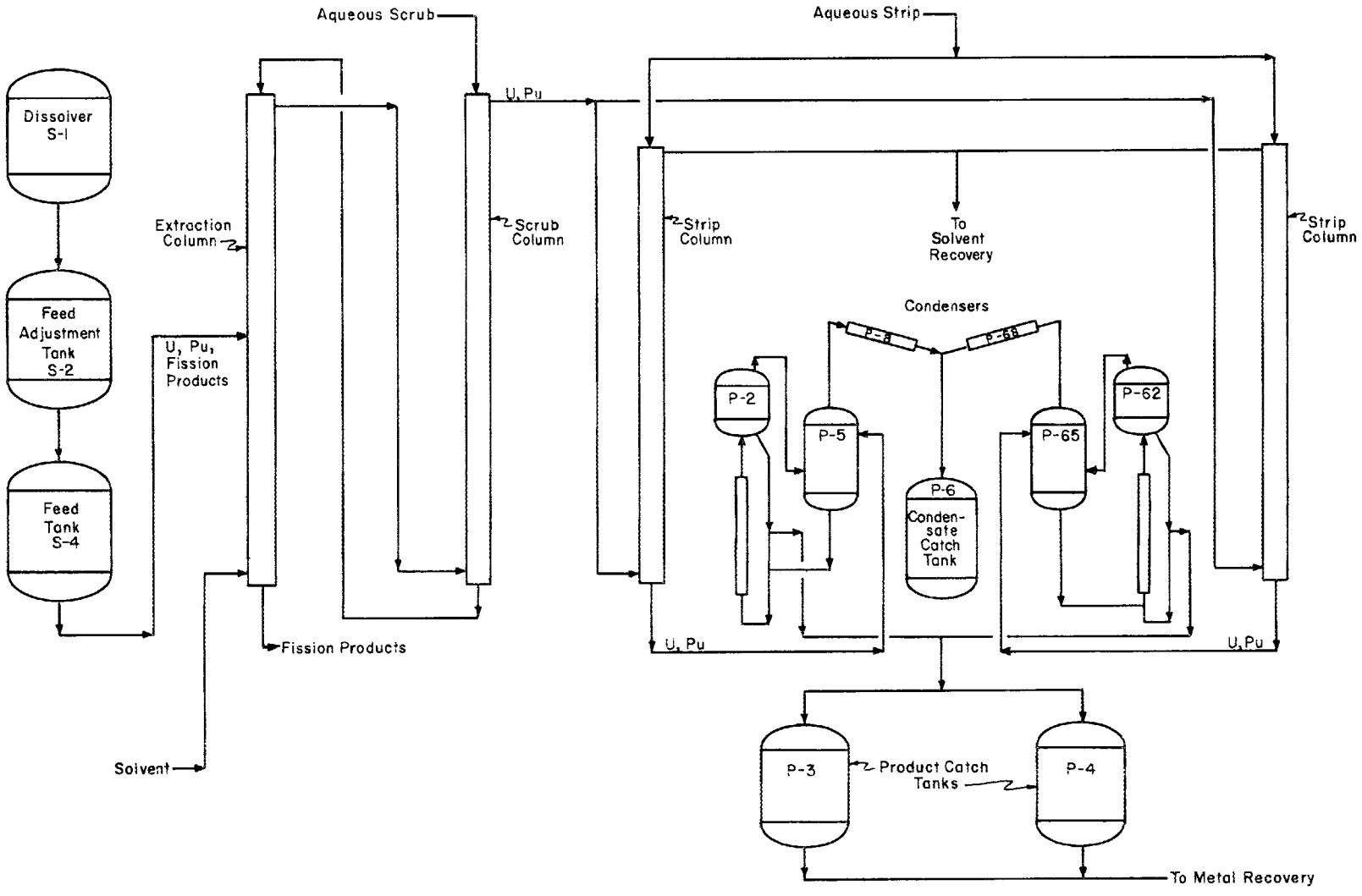

Fig. A.1. PRFP Co-decontamination cycle, 3019 Bldg.

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